Laboratory of Quantum Magnetism

The Neutron Scattering Structure Factor

Physics Master - Travaux Pratiques TPIV

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Contents

1 Introduction

In the experience of neutron scattering, the structure factor is a value that can be almost directly measured (through the differential inelastic neutron scattering cross section). Thus it can be interesting to have a calculation which could give a theoretical point of view, even if the system used in the calculation cannot be as complex as in the real case. Although this calculation (of a simplified system) could be a bad representation of the real system, it is already a point of comparison which yields finally to a better understanding.

The present work has the aim to provide a useful matlab code for the computation of the structure factor of small magnetic cluster in "spin-only" scattering. A concrete exemple and the result for a special configuration will be also given.

2 Structure factor

Before coming to the stucture factor, we have first to define the neutron cross section. In the chapter 1 of the "Theory of neutron scattering from condensed matter" [1], they give the differential cross section in the case of elastic scattering :

$$\frac{d\sigma}{d\Omega} = |\langle\vec{k'}|\hat{V}|\vec{k}\rangle|^2 \tag{1}$$

$\hat{V}$ is the interaction potential that causes the transition from the state $|\vec{k}\rangle$ of the neutron to the state $|\vec{k'}\rangle$ at the same energy. In our case the interaction between the incident neutron and the target sample. We can now include inelastic scattering events, thus we have to take into account that the neutron energy changes. This change is taken in the response of the target sample through a rearrangement of its states. We have to introduce the eigenstate of the target $|\lambda\rangle$ and their energies associated E_λ. The energy conservation leads to :

$$\hbar\omega = E_{\lambda'} - E_\lambda \tag{2}$$

It means that the possible excitations are discretes. With that and the fact that the states of the system are composed of the incident neutron and target (described by a product state fuction $|\vec{k}\rangle|\lambda\rangle = |\vec{k}\lambda\rangle$), we can give the partial differential cross-section :

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)^\lambda_{\lambda'} = \frac{k'}{k}|\langle\vec{k'}\lambda'|\hat{V}|\vec{k}\lambda\rangle|^2\delta(\hbar\omega + E_\lambda - E_{\lambda'}) \tag{3}$$

The factor (k'/k) comes from the density of final neutron states divided by the incident neutron flux. The δ function represent here the energy conservation. By association of a (normalized) weight p_λ with every state $|\lambda\rangle$, we obtain the Born approximation :

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k}\sum_{\lambda\lambda'}p_\lambda|\langle\vec{k'}\lambda'|\hat{V}|\vec{k}\lambda\rangle|^2\delta(\hbar\omega + E_\lambda - E_{\lambda'}) \tag{4}$$

This value gives the fraction of neutrons of incident energy E scattered into an element of solid angle $d\Omega$ with an energy between E' and $E' + dE'$. We will now focus our attention on magnetic scattering. Thus we are interested by the interaction of the neutrons with the magnetic field of the target sample. Again in the reference [1] in the chapter 7, the partial differntial cross-section is given after integration on the neutron coordinates by :

$$\frac{d^2\sigma}{d\Omega dE'} = r_0^2\frac{k'}{k}\sum_{\alpha\beta}(\delta_{\alpha\beta} - \tilde{q}_\alpha\tilde{q}_\beta)\sum_{\lambda\lambda'}p_\lambda\langle\lambda|\hat{Q}_\alpha^\dagger|\lambda'\rangle\langle\lambda'|\hat{Q}_\beta|\lambda\rangle\delta(\hbar\omega + E_\lambda - E_{\lambda'}) \tag{5}$$

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We consider here unpolarized neutrons, r_0 is the classical electron radius and the identity $(\delta_{\alpha\beta} - \tilde{q}_\alpha \tilde{q}_\beta)\hat{Q}_\alpha^\dagger \hat{Q}_\beta$ corresponds to the magnetic interaction. We have also $\tilde{q} = \vec{q}/ \mid q \mid$ with $\vec{q}$: the scattering vector. As we are just interested in spin-only scattering [2], we can write the interaction as follows (neglecting the orbital part) :

$$\langle \lambda' | \hat{Q} | \lambda \rangle = \sum_l exp(i\vec{q} \cdot \vec{R_l}) F(\vec{q}) \langle \lambda' | \hat{S_l} | \lambda \rangle \tag{6}$$

$\hat{S_l}$ is the spin operator of site l, $\vec{R_l}$ is the position of one of the sites in the cristal considered as our target and $F(\vec{q})$ is the ionic form factor. Thus we can obtain the partial differential cross-section for the magnetic scattering by ions with only spin angular momentum :

$$\frac{d^2\sigma}{d\Omega dE'} = r_0^2 \frac{k'}{k} \mid F(\vec{q}) \mid^2 \sum_{\alpha,\beta}(\delta_{\alpha\beta} - \hat{q}_\alpha \hat{q}_\beta) S_{\beta\alpha} \tag{7}$$

Where $S_{\beta\alpha}$ is the structure factor given by :

$$S_{\beta\alpha} = \sum_{\lambda,\lambda'} p_\lambda \sum_{l,d} exp(i\vec{q} \cdot (\vec{R_d} - \vec{R_l})) \langle \lambda | S_l^\alpha | \lambda' \rangle \langle \lambda' | S_d^\beta | \lambda \rangle \delta(\hbar\omega + E_\lambda - E_{\lambda'}) \tag{8}$$

The structure factor is the central element of this work, because it contains in itself all the informations about the interactions, the structure and the magnetic order of the target sample that we would like to study. It is a link between the properties of the microscopic system and our observations with the help of neutron probes. We can still specilise the structure factor in an "exclusive structure factor", because the function $\delta(\hbar\omega + E_\lambda - E_{\lambda'})$ imposes discrete levels. The exclusive structure factor is a computation within specific magnetic multiplets : $|\lambda_i\rangle$ at energy E_i and $|\lambda'_f\rangle$ at energy E_f. This yields to the following expression :

$$S_{\beta\alpha}^{(fi)} = \sum_{\lambda,\lambda'} p_\lambda \sum_{l,d} exp(i\vec{q} \cdot (\vec{R_d} - \vec{R_l})) \langle \lambda_i | S_l^\alpha | \lambda'_f \rangle \langle \lambda'_f | S_d^\beta | \lambda \rangle \tag{9}$$

We can see through this that the exclusive differential inelastic neutron scattering cross section is proportional to the exclusive structure factor in the following way (taken the form factor $F(\vec{q})$, k and k' as constants) :

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$$\frac{d^2\sigma^{(fi)}}{d\Omega} \propto (\delta_{\alpha\beta} - \hat{q}_\alpha \hat{q}_\beta) S_{\beta\alpha}^{(fi)} \tag{10}$$

The second remark about the structure factor is that we cannot compute it without the eigenstates $|\lambda\rangle$ of the system. The system in our calculation is a small magnetic cluster of spin $\frac{1}{2}$. At this point we define the interactions between the spins of our cluster (the "plaquette" of our system) and then compute the Heisenberg Hamiltonian associated :

$$H = \sum_{(i,j)} J_{ij} \vec{S}_i \cdot \vec{S}_j \tag{11}$$

Where J_{ij} corresponds to the interaction between the spin at site i and the spin at site j. By diagonalisation on a convenient basis, more precisely on the S^z basis, we find the eigenvalues and the eigenstates which will be used to compute the structure factor.

For neutron experiments on powder samples, it is also useful to compute an average of the structure factor on all the directions. Thus we have :

$$\bar{S}(q) = \int \frac{d\Omega_{\hat{q}}}{4\pi} S(\vec{q}) \tag{12}$$

3   User's guide

In this guide we first describe the different functions that contain the matlab code which compute the structure factor and then we give an example of the simple dimer calculation.

3.1   The functions

Initially we give the functions which lead to the operators hamiltonian, S_Z^{tot} and S_{tot}^2 in the S_i^z basis. After We need to diagonalise the system to find its eigenstates and eigenvalues, for this purpose we have the function **Qeig**. Then we have also a funtion which displays the spin and eigenvalue according to every eigenstate. Finally we have the functions which give the structure factor.

Si(i,N) :

Gives the spin $\frac{1}{2}$ operator for the site i. It is defined by the following tensor product :

$$S_i^a = \overbrace{\underbrace{Id \otimes Id \otimes \ldots \otimes Id}_{i-1} \otimes S^a \otimes Id \otimes \ldots \otimes Id}^{N} \qquad (13)$$

Id is the identity tensor, S^a corresponds to the pauli matrices divided by 2 ($\hbar = 1$ in this calculation) and N the total number of sites.

SiSj(i,j,N) :

Uses the function **Si(i,N)** to create the operators of the site i and of the site j, then multiply these two operators by each other and returns the result.

sumSiSj(plaquette, Js) :

Computes the hamiltonien (equation 11) following the different interactions between the sites of the cluster used. "plaquette" is a matrix which gives according to its indices (i,j) the interaction (more precisely the element in Js) between the site i and the site j.

Sztot(N) :

Returns for N spins $\frac{1}{2}$ the operator S_Z^{tot}, which is simply the sum of the z component of the spin operator over all the sites.

Stotsq(N) :

Returns for N spins $\frac{1}{2}$ the operator S_{tot}^2 :

$$S_{tot}^2 = \sum_i \sum_j \vec{S}_i \cdot \vec{S}_j \tag{14}$$

Qeig(H,A) :

Diagonalizes the operator H with the function "eig" of matlab, then it projects A on the different eigen-subspace of H and diagonlizes again these projections, which leads also to the complete diagonalization of A. At the end the function returns the eigenstates and eigenvalues associated.

affichage(St,H,eval,evec) :

Takes in argument the operator S_{tot}^2, the hamiltonian, the eigenvalues and the eigenvectors. It shows the energy, the spin and the state of every eigenstate. It uses the functions **ket(i,N)** and **state2str(state)** to show the state in a readable manner and the function **spincal(St,eval,evec)** to compute the spin for every eigenstate.

Structfac(sites,gs,ex,v,t,qx,qy,qz) :

Computes the structure factor $S_{vt}^{(gs-ex)}$ as it is given in the precedent section (equation 9). "sites" is the configuration of the cluster used, gs and ex select between which states the computation is done, (v,t) gives the indices of the structure factor tensor and finally qx,qy and qz are the components of the vector $\vec{q}$. The code allows to put directly array instead of value for qx, qy and qz. The function returns the structure factor.

PowderSF(sites,gs,ex,v,t,r) :

Computes the structure factor powder average by using the function **Structfac(sites,gs,ex,v,t,qx,qy,qz)**. The value r corresponds to the norm of $\vec{q}$. More in details, we take an amount of points (about 200'000) uniformly distributed on a sphere, they represent the orientation of the

vector $\vec{q}$, then we compute the structure factor for all these orientations. The average is taken by summing all these structure factor divideb by the number of contributions.

3.2 Example of a code

We give here the example of a calculation on a spin dimer to see how it works

```
% Defines geometry and topology of the cluster
sites=[1 0 0; 0 1 0];
plaquette=[0,1;1,0];
J=1;
N=size(plaquette,1);

% Computes the operators in the S^z basis
H=sumSiSj(plaquette,J);
St=Stotsq(N);
Sz=Sztot(N);

% Diagonalizes simultaneously H and St
[evec eval]=Qeig(H,St);

% Output eigenstates
affichage(St,H,eval,evec);

% q-space
q=[0:0.002:1]*6;
[qx,qy]=meshgrid(q,q);
qz=zeros(size(qx));

% Selects ground state and excitation
gs=evec(:,1);
ex=evec(:,2:4);

% Computes the structure factor
StructF=Structfac(sites,gs,ex,1,1,2*pi*qx,2*pi*qy,2*pi*qz);

% Plots results
figure(1)
p=pcolor(qx,qy,abs(StructF));
set(p,'LineStyle','none');
title('Structure_Factor');
xlabel('q_h[rlu]');
ylabel('q_k[rlu]');
```

```
Analytic=(1−cos(2∗pi∗q))/2;

figure(2)
hold on;
h1=plot(q,Analytic,'−','linewidth',1.2);
h2=plot(q,real(StructF(1,:)),'xr','linewidth',1.2);
hold off;
title('Structure Factor');
xlabel('q_h (q_k=0)');
ylabel('S_{11}^{GS−1^{st}Ex}');

% Powder average
r=(0:0.1:15);
pwSF=PowderSF(sites,gs,ex,1,1,r);

figure(3)
h3=plot(r,pwSF,'−', 'linewidth',1.2);
xlabel('r','FontSize',20);
ylabel('S(q)','FontSize',20);
title({'Stucture factor : Powder average'},'FontSize',24);
```

First we define our system with **sites**, **plaquette** and **J**, they are the positions of copper atoms and their interactions. Every line of **sites** gives the position of one atom in an orthogonal system. **plaquette** is a symmetric square matrix, where every element (i,j) gives where in the array **J** is the interaction between the site i and the site j. Secondly we compute some operators : the hamiltonian, S_Z^{tot} and S_{tot}^2. Then we use the function **Qeig** to calculate the eigenvalues and eigenstates, that we display in the matlab terminal with the function **affichage**. we define also an array of vector $\vec{q}$ with meshgrid and plot the structure factor $S_{11}^{gs->ex}$ in a color plot in function of q_h and q_k (see figure 1). "gs" is the ground state (singlet) and "ex" is an excited multiplet (triplet). The second plot (figure 3) gives the structure factor only in function of qh ($qk = 0$) compared to the analytical solution given below :

$$S_{11}^{GS-1^{st}Ex} = \frac{1}{2}(1 - cos(\vec{q}\cdot\vec{a})) \qquad (15)$$

The vector $\vec{a}$ represents the diffence between the positions of the dimer. We can see on figure 3 that our calcultion matches very well with the analytic solution. Finaly the last plot (figure 2) shows the structure factor for a powder average.
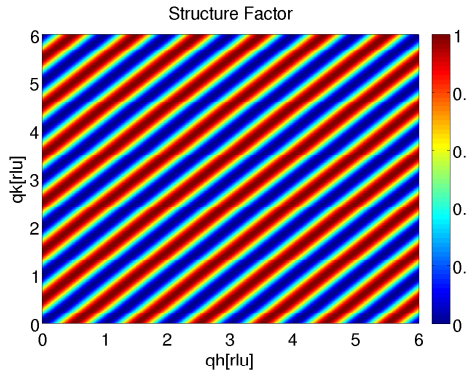
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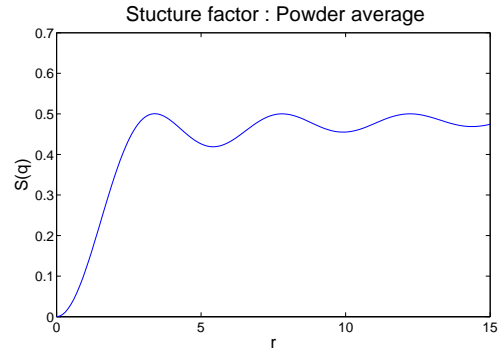Figure 1: *Color plot of the structure factor $S_{11}^{gs->ex}$*

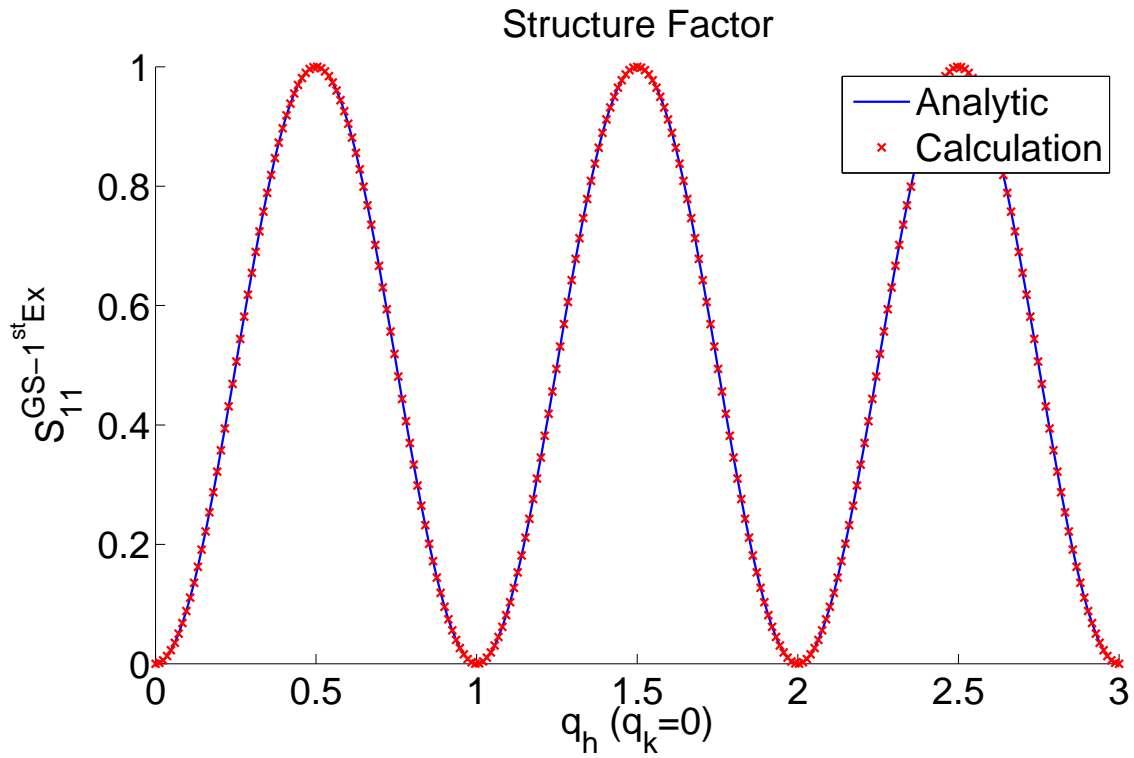Figure 2: *This figure shows the powder average structure factor of a spin dimer.*

Figure 3: *The structure factor $S_{11}^{gs->ex}$ in function of q_h ($q_k = 0$).*

The color plot, like in the figure 1, is a nice way to represent the structure factor in function of (q_x, q_y). We can see directly for exemple that there is a symmetry in the structure factor $(q_x \leftrightarrow q_y)$, which can be explain by the symmetry of the system (where are placed the ion copper sites) we have taken.

4   Result : Phase transition in quadrimer
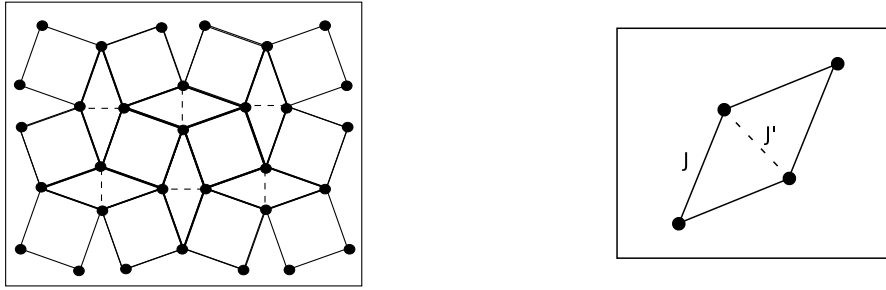
We study here the configuration of $SrCu_2(BO_3)_2$ (SCBO). We suppose that it is a model with an intermediate phase between the dimer and the antiferromagnetic phases [4]. The figures above show a model where the limit cases ($J/J' << 1$ and $J/J' >> 1$) correspond to the dimer and to the antiferromagnetic phases. The problem now is that we cannot take this whole system for problem of computation (it would take to much time to compute it), thus we approximate this system to an easier one : a quadrimer with diagonal interaction as it is also shown above. We compute the neutron scattering structure factor of this cluster and compare with two dimers in the same configuration.

$$J = 10 \cdot J' \tag{16}$$

The factor between J and J' is not the real one, but the important point is that J is bigger than J'. In fact we do not use the exact value because it is not really helpful in our approximation. When we compute the structure factor on the SCBO cluster it gives only a first approximation of the reallity, first because it is only a tiny part of the whole system, then because the interactions are more complex, for exemple we didn't take into account the correlations with the other plaquettes.

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M. Zayed in the chapter 5 of his thesis [3] analyses quantum phase transition in SCBO induced by pressure. He pointed out 3 characteristics of the high pressure phase:

- a non magnetic gapped ground state

- a low energy triplet with structure factor peaking at Q=(1,0)

- an other low energy excitation with structure factor indentical to the singlet-triplet in the exact dimer phase.

The following tabular shows the energie and the spin of every eigenstate of the plaquette and of the 2 dimers in the SCBO configuration. We can see that the ground state in both case is isolated and has a spin equal to zero. For the dimers, the first excited multiplet corresponds to two triplet. And for the plaquette, the first excited multiplet is a triplet and the second one is a quadruplet. The figure 4 gives the differential inelastic neutron scattering cross section using the structure factor. We can see that the transition between the ground state and the second excited multiplet for the plaquette corresponds to the transition for the dimers. The other transition for the plaquette (GS → 1st excitation) is different, but it has a peak around $q_x = 1$. Even though our results stay an approximation, they confirm these three characteristics. Thus we are on the good way to say that the plaquette with the diagonal bond is the best candidate for the SCBO configuration.

	Plaquette SCBO		2 dimers	
	Energy	Spin	Energy	Spin
1	−1.9750	0	−1.5000	0
2	−0.9750	1	−0.5000	1
3	−0.9750	1	−0.5000	1
4	−0.9750	1	−0.5000	1
5	−0.0750	1	−0.5000	1
6	−0.0750	0	−0.5000	1
7	−0.0750	1	−0.5000	1
8	−0.0750	1	0.5000	0
9	0.0250	1	0.5000	2
10	0.0250	1	0.5000	2
11	0.0250	1	0.5000	2
12	1.0250	2	0.5000	1
13	1.0250	2	0.5000	1
14	1.0250	2	0.5000	1
15	1.0250	2	0.5000	2
16	1.0250	2	0.5000	2

5   Conclusion

The neutron scattering structure factor is a very important quantity to interpret experiments with neutron scattering. The exemple of phase transition in SCBO shows in a way the possibility of the structure factor code. Finally I hope that we created a useful tool for those who need it.
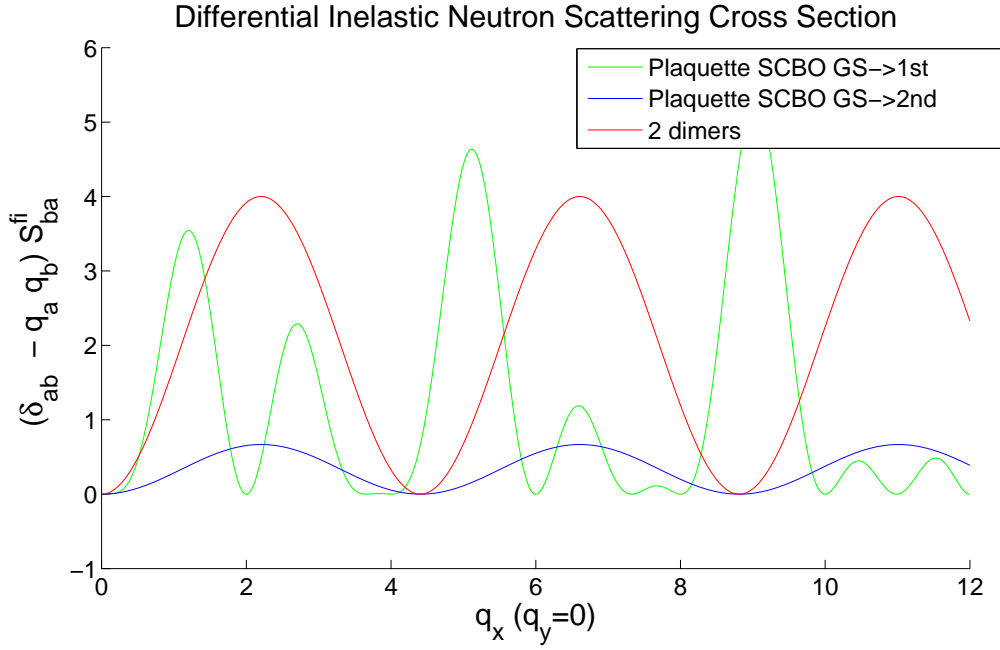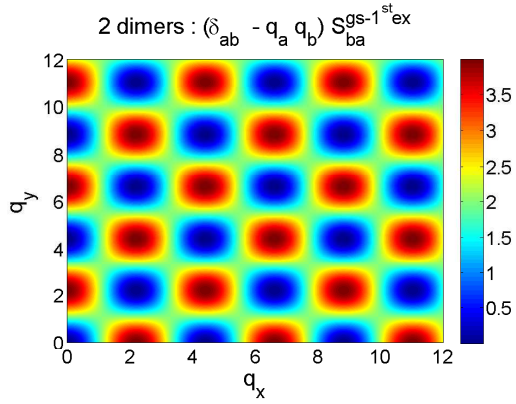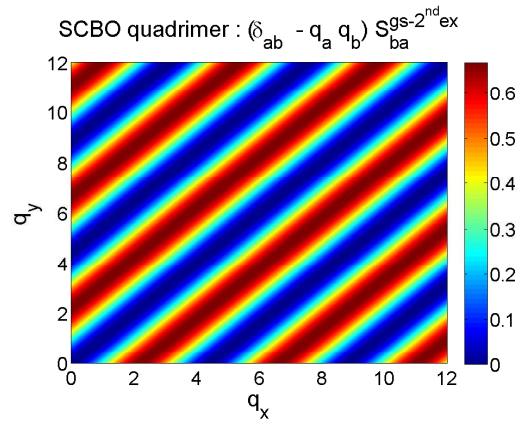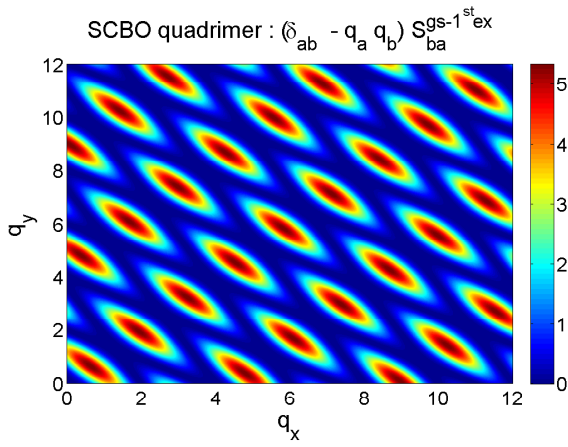
6   Acknowledgments

Figure 4: *This graph shows the differential inelastic neutron scattering cross section in function of q_x. The 3 curves represent different excitations and configurations in the SCBO geometry.*

References

[1] S. W. Lovesey, Theory of neutron scattering from condensed matter, vol. 1 and 2, Oxford University Press, 1984

[2] J. T. Haraldsen et al., Phy. Rev. B. **71**, 064403 (2005)

[3] M. Zayed, Thesis, Ch. 5 "Pressure induced quantum phases" (2010)

[4] M. Albrecht and F. Mila, Europhys. Lett. **34**, 145 (1996)

SCBO quadrimer : $(\delta_{ab} - q_a q_b) S_{ba}^{gs-1^{st}ex}$

SCBO quadrimer : $(\delta_{ab} - q_a q_b) S_{ba}^{gs-2^{nd}ex}$

2 dimers : $(\delta_{ab} - q_a q_b) S_{ba}^{gs-1^{st}ex}$

These color plots are associated with the figure 4, they give a more general view of the differential inelastic neutron scattering cross section.

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