Neutron and X-ray Scattering of Quantum Materials

PHYS-640

Week 7 exercises

1: The energy resolution in a backscattering experiment

The uncertainty on the final energy of the detected neutron is governed by many factors but one large factor is the scattering angle itself. Here we have a closer look at why that is.

(a) Use Bragg's law, $n\lambda_f = 2d_{\text{ana}} \sin\theta_f$, and propagation of errors to find an expression for the relative uncertainty $\frac{\delta\lambda_f}{\lambda_f}$. Here λ_f is the final wavelength of the neutron, d_{ana} is the lattice spacing of the monochromator and $2\theta_f$ is the scattering angle of the neutrons arriving at the detector.

Propagating the error gives:

$$\begin{aligned}(\delta\lambda_f)^2 &= \left(\frac{\partial(2d_{\text{ana}}\sin\theta_f)}{\partial d_{\text{ana}}}\right)^2 (\delta d_{\text{ana}})^2 + \left(\frac{\partial(2d_{\text{ana}}\sin\theta_f)}{\partial\theta_f}\right)^2 (\delta\theta_f)^2 \\ &= (2\sin\theta_f)^2\,(\delta d_{\text{ana}})^2 + (2d_{\text{ana}}\cos\theta_f)^2(\delta\theta_f)^2.\end{aligned}$$

The relative uncertainty is then:

$$\left(\frac{\delta\lambda_f}{\lambda_f}\right)^2 = \left(\frac{\delta d_{\text{ana}}}{d_{\text{ana}}}\right)^2 + (\cot\theta_f)^2(\delta\theta_f)^2.$$

(b) At which scattering angle is $\frac{\delta\lambda_f}{\lambda_f}$ at a minimum and what does that mean for the detector position if you wish a very high energy resolution?

The term $\left(\frac{\delta d_{\text{ana}}}{d_{\text{ana}}}\right)^2$ refers to the quality of the analyzer crystal and does not depend on the scattering angle. The other term, $(\cot\theta_f)^2$, has a minimum at $\theta_f = 90°$ which means a scattering angle of $2\theta_f = 180°$. That means scattering back the way the neutrons came and is known as backscattering.

2: The Heisenberg ferromagnetic chain

Consider a Heisenberg ferromagnetic chain with the following spin Hamiltonian:

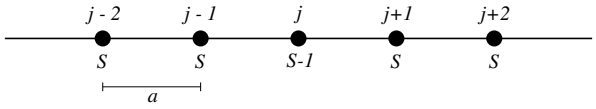
$$\hat{\mathcal{H}} = \sum_i J\,\mathbf{S}_i \cdot \mathbf{S}_{i+1},$$

where the sum is only over the nearest neighbors distanced a from each other, see Fig. 1(a), and we assume that all spin pairs interact with the same exchange coupling, J.

(a) Use the operators $S^+ = S^x + iS^y$ and $S^- = S^x - iS^y$ to arrive at the following form for the Hamiltonian:

$$\hat{\mathcal{H}} = \sum_i J\left[S_i^z S_{i+1}^z + \frac{1}{2}\left(S_i^+ S_{i+1}^- + S_i^- S_{i+1}^+\right)\right]$$

(a)



(b)

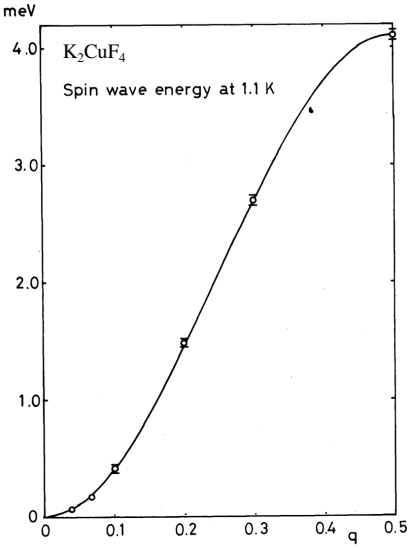


Figure 1: (a) The ferromagnetic chain with spins separated by the distance a and interacting with exhange coupling J. (b) Spinwave spectrum of K_2CuF_4 measured by K. Hirakawa *et al.*, *J. Phys. Soc. Jpn.* **52**, 4220-4230 (1983).

Using the expressions for S^+ and S^- we can write S^x and S^y as:

$$S^x = \frac{1}{2}\left(S^+ + S^-\right), \qquad S^y = \frac{1}{2i}\left(S^+ - S^-\right).$$

Plugging that into the Hamiltonian then yields:

$$\begin{aligned}
\hat{\mathcal{H}} &= \sum_i J\left[S_i^z S_{i+1}^z + S_i^x S_{i+1}^x + S_i^y S_{i+1}^y\right] \\
&= \sum_i J\left[S_i^z S_{i+1}^z + \frac{1}{4}\left(S_i^+ + S_i^-\right)\left(S_{i+1}^+ + S_{i+1}^-\right) - \frac{1}{4}\left(S_i^+ - S_i^-\right)\left(S_{i+1}^+ - S_{i+1}^-\right)\right] \\
&= \sum_i J\left[S_i^z S_{i+1}^z + \frac{1}{4}\left(S_i^+ S_{i+1}^+ + S_i^+ S_{i+1}^- + S_i^- S_{i+1}^+ + S_i^- S_{i+1}^-\right)\right. \\
&\qquad\qquad \left. - \frac{1}{4}\left(S_i^+ S_{i+1}^+ - S_i^+ S_{i+1}^- - S_i^- S_{i+1}^+ + S_i^- S_{i+1}^-\right)\right] \\
&= \sum_i J\left[S_i^z S_{i+1}^z + \frac{1}{2}\left(S_i^+ S_{i+1}^- + S_i^- S_{i+1}^+\right)\right].
\end{aligned}$$

(b) The operators S_j^+ and S_j^- have the effect of respectively raising and lowering the spin value at a site j. S_j^z works a bit like a number operator and measures the spin value on site j. Argue that the state with all spins at their maximum value, S, is an eigenstate of the Hamiltonian and determine the eigenvalue.

The term with $S_i^z S_{i+1}^z$ operating on a state with all spins up gives S^2 for all pairs of i and $i+1$ and leaves the state unchanged. The mixed terms, $S_i^+ S_{i+1}^-$ and $S_i^- S_{i+1}^+$, are attempting to raise the spins on sites i and $i+1$, respectively, but since all spins already attain their maximum value, these terms give zero per definition. Therefore, $\hat{\mathcal{H}}\left|S,S,S,...,S\right\rangle = NJS^2\left|S,S,S,...,S\right\rangle$ and therefore $\left|S,S,S,...,S\right\rangle$ is an eigenstate of the Hamiltonian with eigenvalue NJS^2.

(c) Let us now consider a state with all spins at their maximum except the one at j which is now lowered to $S-1$, meaning $\left|j\right\rangle = \left|S,S-1,S,...,S\right\rangle$. The operators S_j^+ and S_j^- do the following to the state $\left|j\right\rangle$:

$$S^+\left|j\right\rangle = \sqrt{2S}\left|j+1\right\rangle, \qquad S^-\left|j\right\rangle = \sqrt{2S}\left|j-1\right\rangle$$

Is $|j\rangle$ an eigenstate of $\hat{\mathcal{H}}$?

To answer the question we need to evaluate $\hat{\mathcal{H}}\,|j\rangle$. From question (b) we already know that the $S^{\pm}$ part of the Hamiltonian only gives non-zero contributions to the sum when the lowered spin is involved in a pair. That mean we have to look at the pairs $(j-1,j)$ and $(j,j+1)$:

$$\begin{aligned}
S^+_{j-1}S^-_j\,|j\rangle &= S^+_{j-1}\sqrt{2S}\,|S,S-2,S,...,S\rangle = 0,\\
S^-_{j-1}S^+_j\,|j\rangle &= S^-_{j-1}\sqrt{2S}\,|S,S,S,...,S\rangle = 2S\,|j-1\rangle\,,\\
S^+_jS^-_{j+1}\,|j\rangle &= S^+_j\sqrt{2S}\,|S,S-1,S-1,...,S\rangle = 2S\,|j+1\rangle\,,\\
S^-_jS^+_{j+1}\,|j\rangle &= 0.
\end{aligned}$$

The effect of $S^z_iS^z_{i+1}$ part of the Hamiltonian on $|j\rangle$ is evaluated as follows:

$$\begin{aligned}
S^z_{j-1}S^z_j\,|j\rangle &= S^z_{j-1}(S-1)\,|j\rangle = S(S-1)\,|j\rangle\,,\\
S^z_jS^z_{j+1}\,|j\rangle &= S^z_jS\,|j\rangle = (S-1)S\,|j\rangle\,.
\end{aligned}$$

All remaining $S^z_iS^z_{i+1}$ pairs yield S^2. In other words, this part of the sum gives us S^2 for the $N-2$ pairs not involving the lowered spin and $S(S-1)$ for the 2 pairs that do. Putting it all together we have:

$$\begin{aligned}
\hat{\mathcal{H}}\,|j\rangle &= \sum_i J\left[S^z_iS^z_{i+1} + \frac{1}{2}\left(S^+_iS^-_{i+1} + S^-_iS^+_{i+1}\right)\right]|j\rangle\,,\\
&= J\Big[(N-2)S^2 + 2S(S-1)\Big]\,|j\rangle + 2JS\,|j-1\rangle + 2JS\,|j+1\rangle\,,\\
&= J\left(NS^2-2S\right)|j\rangle + 2JS\,|j-1\rangle + 2JS\,|j+1\rangle\,.
\end{aligned}$$

So $|j\rangle$ is not an eigenstate of $\hat{\mathcal{H}}$.

(d) Hopefully you found that a spin lowered on a single site is not an eigenstate of $\hat{\mathcal{H}}$. Instead we try the Fourier transform of $|j\rangle$:

$$|q\rangle = \frac{1}{\sqrt{N}}\sum_j e^{i\mathbf{q}\cdot\mathbf{r}_j}\,|j\rangle\,.$$

We can also view this as a smearing of the lowered spin over all lattice sites. Evaluate $\hat{\mathcal{H}}\,|q\rangle$ and show that this is indeed an eigenstate of the Hamiltonian with eigenvalue $E = JNS^2 + 2JS\,[\cos(qa)-1]$.

To evaluate $\hat{\mathcal{H}}\,|q\rangle$ we can use the result from question (c):

$$\begin{aligned}
\hat{\mathcal{H}}\,|q\rangle &= \frac{1}{\sqrt{N}}\sum_j e^{i\mathbf{q}\cdot\mathbf{r}_j}\hat{\mathcal{H}}\,|j\rangle\,,\\
&= \frac{JS}{\sqrt{N}}\sum_j e^{i\mathbf{q}\cdot\mathbf{r}_j}\Big[\,(NS-2)\,|j\rangle + 2\,|j-1\rangle + 2\,|j+1\rangle\,\Big].
\end{aligned}$$

To go on, we write out the sum and rearrange the terms:

$$\begin{aligned}
\hat{\mathcal{H}}\,|q\rangle = \frac{JS}{\sqrt{N}}\Big(&... + e^{-i2qa}\left[(NS-2)\,|j-2\rangle + 2\,|j-3\rangle + 2\,|j-1\rangle\,\right]\\
&+ e^{-iqa}\left[(NS-2)\,|j-1\rangle + 2\,|j-2\rangle + 2\,|j\rangle\,\right]\\
&+ \left[(NS-2)\,|j\rangle + 2\,|j-1\rangle + 2\,|j+1\rangle\,\right]\\
&+ e^{iqa}\left[(NS-2)\,|j+1\rangle + 2\,|j\rangle + 2\,|j+2\rangle\,\right]\\
&+ e^{i2qa}\left[(NS-2)\,|j+2\rangle + 2\,|j+1\rangle + 2\,|j+3\rangle\,\right] + ...\Big),
\end{aligned}$$

$$
\begin{aligned}
\hat{\mathcal{H}}\,|q\rangle &= \frac{JS}{\sqrt{N}}\Big[\ldots + \left(2e^{-i2qa} + e^{-iqa}(NS-2) + 2\right)|j-1\rangle \\
&\qquad + \left(2e^{-iqa} + (NS-2) + 2e^{iqa}\right)|j\rangle \\
&\qquad + \left(2 + e^{iqa}(NS-2) + 2e^{i2qa}\right)|j+1\rangle + \ldots\Big), \\
&= \frac{JS}{\sqrt{N}}\left[\sum_j (NS-2)e^{i\mathbf{q}\cdot\mathbf{r}_j}\,|j\rangle\right] + \frac{JS}{\sqrt{N}}\Big[\ldots + \left(2e^{-i2qa}+2\right)|j-1\rangle \\
&= \qquad\qquad + \left(2e^{-iqa} + 2e^{iqa}\right)|j\rangle + + \left(2 + 2e^{i2qa}\right)|j+1\rangle + \ldots\Big], \\
&= JS(NS-2)\,|q\rangle + \frac{JS}{\sqrt{N}}\Big[\ldots + 2e^{-iqa}\left(e^{-iqa} + e^{iqa}\right)|j-1\rangle + 2\left(e^{-iqa} + e^{iqa}\right)|j\rangle \\
&= \qquad\qquad +2e^{iqa}\left(e^{-iqa} + e^{iqa}\right)|j+1\rangle + \ldots\Big], \\
&= JS(NS-2)\,|q\rangle + \frac{4JS}{\sqrt{N}}\sum_j \cos(qa)e^{i\mathbf{q}\cdot\mathbf{r}_j}\,|j\rangle\,, \\
&= JS\Big[(NS-2) + 2\cos(qa)\Big]\,|q\rangle\,, \\
&= \Big[JNS^2 + 2JS\left(\cos(qa) - 1\right)\Big]\,|q\rangle\,.
\end{aligned}
$$

This means that $|q\rangle$ is in fact an eigenstate of $\hat{\mathcal{H}}$ with eigenvalue $E = JNS^2 + 2JS\left(\cos(qa) - 1\right)$.

(e) Plot the spinwave spectrum for a ferromagnetic chain with $S = 2$ and $J = -5$ meV.

The constant part of E is basically the ground state energy and does not contribute to the dispersion. With the given parameters, the dispersion is shown in Fig. 2.

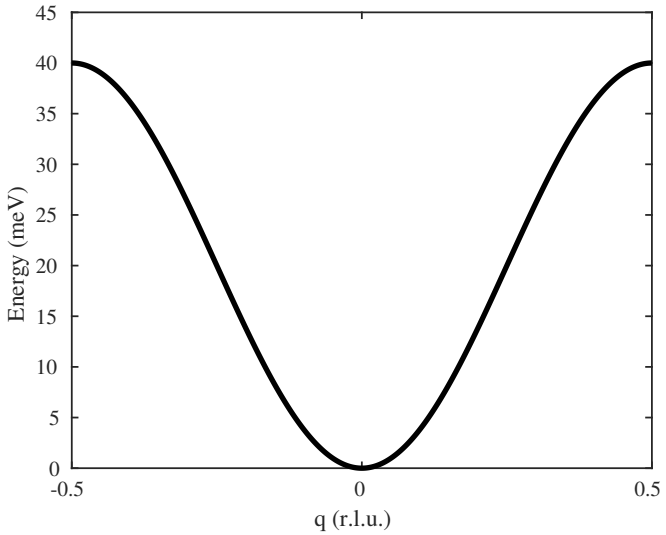


Figure 2: The spinwave dispersion for a ferromagnetic chain with $S = 2$ and $J = -5$ meV.

(f) The spinwave spectrum of the ferromagnetic chain compound, K_2CuF_4, is shown in Fig. 1(b). What is the value of J in this case?

To determine the height of the dispersion (also known as the bandwidth) we need to find the maximum of $E(q)$:

$$
\frac{dE(q)}{dq} = -2JSa\sin(qa) = 0 \quad \Rightarrow \quad qa = 0 \vee qa = \pi.
$$

Here the solution $qa = 0$ is the minimum and $qa = \pi$ is the maximum. The energy at the maximum is then $E_{\max} = -4JS$. (The constant JNS^2 is not regarded as it is just an offset). We read the maximum of the curve in Fig. 1(b) to be around 4 meV. The magnetic ion in this case is Cu^{2+} which has spin $S = \frac{1}{2}$. The exchange interaction is then $J = -\frac{E_{\max}}{4S} = -\frac{4\,\text{meV}}{2} = -2\,\text{meV}$.

3: Lattice vibrations in one dimension

Exercise 14.P.1 in the neutron notes.

1.
The force between ion j and $j+1$ is:

$$F_{j,j+1}(t) = -K\left[u_j(t) - u_{j+1}(t)\right].$$

For the pair $j-1$ and j it is similarly:

$$F_{j-1,j}(t) = -K\left[u_{j-1}(t) - u_j(t)\right].$$

Now $F_{j,j-1} = -F_{j-1,j}$ and with Newton's second law we get the total forces on ion j:

$$M\ddot{u}_j = F_{j,j+1}(t) - F_{j-1,j}(t) = -K\left[2u_j - u_{j-1} - u_{j+1}\right].$$

2.
We plug the trial function $u_j(t) = A_q e^{i(qja-\omega_q t)}$ into the equation of motion (note that $a = d$):

$$\begin{aligned} M(-\omega_q^2)A_q e^{i(qja-\omega_q t)} &= -K\left[2A_q e^{i(qja-\omega_q t)} - A_q e^{i(q(j-1)a-\omega_q t)} + A_q e^{i(q(j+1)a-\omega_q t)}\right] \Leftrightarrow \\ M\omega_q^2 e^{i(qja-\omega_q t)} &= K\left[2e^{i(qja-\omega_q t)} - e^{iqa}e^{i(qja-\omega_q t)} + e^{iqa}e^{i(qja-\omega_q t)}\right] \Leftrightarrow \\ M\omega_q^2 &= K\left[2 - e^{iqa} + e^{iqa}\right]. \end{aligned}$$

Now we use the definition of the sine using the complex exponential, $\sin(x) = \frac{e^{ix}-e^{-ix}}{2i}$, to evaluate $\sin^2(x)$:

$$\sin^2(x) = \left(\frac{e^{ix}-e^{-ix}}{2i}\right)\left(\frac{e^{ix}-e^{-ix}}{2i}\right) = \frac{e^{i2x} - 1 - 1 - e^{-i2x}}{-4} = \frac{2 - e^{i2x} - e^{-i2x}}{4}.$$

We can then substitute $2 - e^{iqa} + e^{iqa}$ by $4\sin^2\left(\frac{qa}{2}\right)$ and we obtain the result:

$$M\omega_q^s = 4K\sin^2\left(\frac{qa}{2}\right) \quad \Leftrightarrow \quad \omega_q^s = \frac{4K}{M}\sin^2\left(\frac{qa}{2}\right).$$

4: The direct-geometry time-of-flight spectrometer

An inelastic neutron scattering experiment is performed with a $CeCu_6$ single crystal on a time-of-flight spectrometer with direct geometry. The space group of $CeCu_6$ is *Pnma* (orthorhombic) with lattice parameters $a = 5.03$ Å, $b = 8.06$ Å and $c = 10.09$ Å. Suppose the sample is oriented with $(H, 0, 0)$ and $(0, 0, L)$ in the horizontal scattering plane. The incoming energy of the neutrons is $E_i = 8$ meV and the detectors cover a range of scattering angles of $10° - 100°$ in the forward direction.

Calculate the $\mathbf{q}$ coverage for an energy transfer of $E = 0.3$ meV when the crystal is rotated by 90° in steps of 1°.

To calculate the coverage we need to use the Laue condition together with conservation of energy:

$$\mathbf{q} = \mathbf{k}_i - \mathbf{k}_f, \qquad \Delta E = E_i - E_f.$$

We also need to be able to convert between energy and momentum with $k = \frac{2m_n E}{\hbar}$, where m_n is the neutron mass and $\hbar$ the reduced Planck constant. To get the units to match, we need the elementary charge, e.

We assume that the incoming neutrons arrive along z such that $\mathbf{k}_i = \frac{2m_n E_i}{\hbar}\hat{\mathbf{z}}$ and we place the sample such that $(H, 0, 0)$ is along x and $(0, 0, L)$ is along z as shown in Fig. 3(a). This is just one choice of scattering geometry. If you want, you can play around with other choices. Figure 3(b) shows the coverage for different values of energy transfer when rotating the crystal from 5° to 95°. Again this is an arbitrary choice and you can try our other choices.

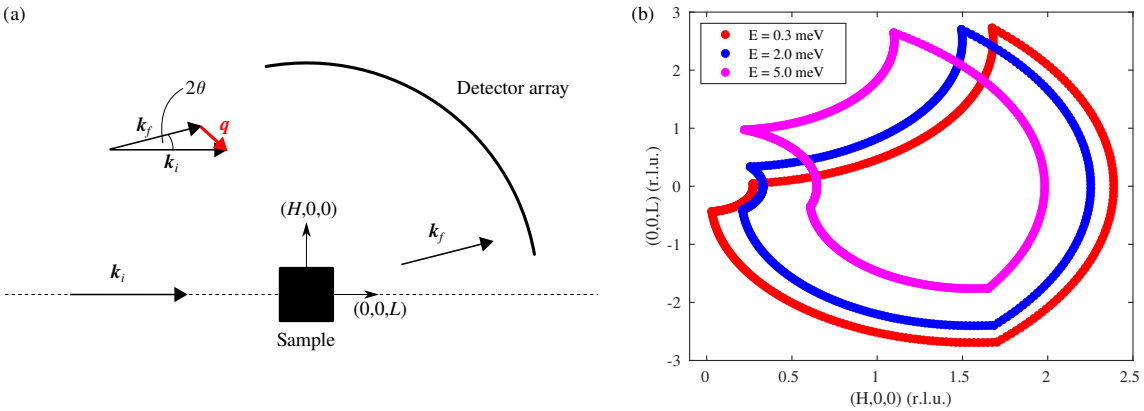


Figure 3: (a) Experimental setup. (b) Coverage in $\mathbf{q}$ for different energy transfers.