Advanced Student Practicum at the
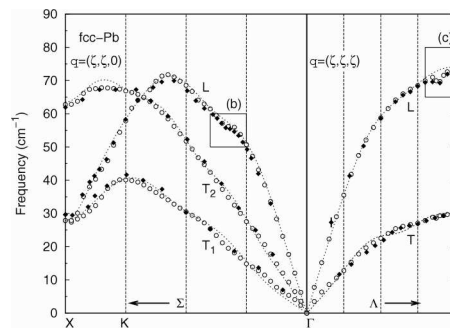*Laboratory for Neutron Scattering (LNS)*
Swiss Spallation Neutron Source (SINQ)
Paul Scherrer Intitut (PSI)

Inelastic Neutron Scattering:

Phonon-dispersions and Lattice Dynamics in a Lead (Pb) Crystal

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Table of Contents

1. Brief Summary of the Practicum

In this practicum you will learn how to conduct an inelastic neutron scattering experiment using a state-of-the-art triple-axis cold neutron spectrometer at the Swiss Neutron Spallation Source (SINQ) of the Paul Scherrer Institut (PSI). Your task is to measure the phonon dispersion curves of a lead single crystal sample and from that deduce a series of fundamental physical properties for this material.
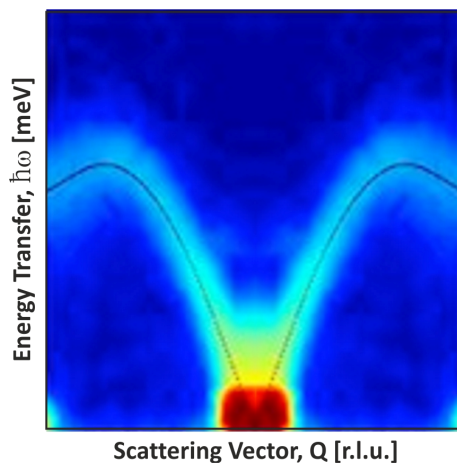
2. Useful References & Properties

General Solid State Physics:

[1]   Ch. Kittel, "Introduction to Solid State Physics"
          John Wiley & Sons; 8th Edition edition (11 Nov 2004)

[2]   N. W. Ashcroft and N. D. Mermin, "Solid State Physics"
          Brooks/Cole (2 Jan 1976)

Neutron Scattering:

[3]   A. Furrer, J. Mesot and Th. Strässle, "Neutron Scattering in Condensed Matter Physics"
          World Scientific Publishing Company; 1st edition (May 22, 2009)

[4]   G. L. Squires, "Introduction to the Theory of Thermal Neutron Scattering"
          Cambridge University Press; 3rd edition (May 7, 2012)

Phonons (in lead):

[5]   P. Brüesch, "Phonons: Theory and Experiments I: Lattice Dynamics and Models…"
          Springer; reprint of 1st ed. 1982 edition (January 4, 2012)

[6]   R. Stedman *et al.*, Physical Review **162**, 545 (1967)

[7]   Andrea Dal Corso, Journal of Physics: Condensed Matter **20**, 445202 (2008)

Lead properties:

Name	Plumbum (Latin for: *liquid silver*)
Symbol	Pb
Atomic Number	82
Atomic Weight	207.2(1) au
Lattice type	fcc
Lattice constants	$\mathbf{a} = \mathbf{b} = \mathbf{c}$ = 5 Å (T = 300 K)
	$\alpha = \beta = \gamma$ = 90°
Molar weight	M = 207.2(1) g/mol
Density	ρ = 11.34 g/cm^3
Brillouin Zone	bcc

Conversion factors for neutrons:

QUANTITY	RELATIONSHIP	VALUE @ E = 10 meV
Energy	[meV] = 2.072·k^2 [Å^{-1}]	10 meV
Wavelength	λ [Å] = 9.044 / $\sqrt{E}$ [meV]	2.86 Å
Wave Vector	k [Å^{-1}] = 2π / λ [Å]	2.20 Å^{-1}
Frequency	ν [THz] = 0.2418·E [meV]	2.418 THz
Wavenumber	ν [cm^{-1}] = ν [Hz] / (2.998·10^{10} cm/s)	80.65 cm^{-1}
Velocity	v [km/s] = 0.6302·k [Å^{-1}]	1.38 km/s
Temperature	T[K] = 11.605·E [meV]	116.05 K

3. Practical Information & Safety Issues

Location:
This laboratory exercise is performed at the Swiss Neutron Spallation Source (SINQ) of the Paul Scherrer Institut (PSI). To find your way here please visit: **http://www.psi.ch/how-to-find-us**

Time Schedule:
The INS practicum runs over 1-1.5 days, starts at 09:30 and will finish at 18:30. There will be plenty of time for breaks/lunch/coffee during the measurements.

Clothing/shoes:
This is a practical exercise and the SINQ facility including the spectrometer area is a large scale laboratory. This means that we will move around and handle experimental equipment that might not be particularly clean or "gentle". Hence, we encourage the students to wear comfortable shoes and try to avoid wearing "fragile" clothing as well as watches or jewelry since there is risk of damage.

Computing:
There a couple of available computers on site (Linux/Windows) that can be used to view/analyze the data. However, we encourage the students to bring their own laptops as well, preferably with some sort of data analysis program installed (e.g. Matlab, Origin, Igor etc.).

SAFETY:

PSI Safety Course is mandatory for users of PSI facilities. Therefore, in order to get a dosimeter and a permission to perform an experiment at PSI, you have to pass the safety course. The most convenient way to pass this course is using the web-link below. Once you passed the course you will be allowed to print a form that you should fill out and sign. This form you should then bring with you on the day of the practicum.

This practicum takes place inside the SINQ experimental building, which is a controlled radiation area. You will be issued a radiation dosimeter that you need to wear at all times. Further, it is very important that you follow the instructions of your supervisor and do not touch anything or enter areas that you are not specifically instructed to use. Further, no eating or drinking is allowed anywhere inside the SINQ building (see pamphlet to the right). Finally, anytime you need to exit the building, you are obliged to first check yourself for contamination using the radiation meters, which are located at the exits. Your supervisor will guide you through this process.

If you follow these instructions the SINQ facility and this practicum should be an interesting and safe experience.

Zone I - SINQ (WNHA/WNLA) + Experimentierhalle

Radiation protection reminder

radiation badge and PC safety course are mandatory in this area

No eating or drinking in the controlled area
including
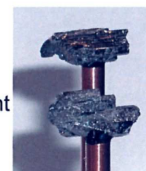- chewing gums
- lip sticks

Handling of **unsealed powder samples**:
Inside the User-Laboratory WLHA only (access on request)

All exposed and unexposed material must be checked by SU safety officer

This includs
- **samples**
- experimental equipment
- tools

Questions ?
Contact a SU safety officer (phone numbers are posted at all exits)

Link to online safety course (choose access to **SINQ** facility):

http://srp.web.psi.ch/su-introduction/Einfuehrung_Experimentatoren_englisch.pps

4. Lattice Vibrations: Phonons

For most instances the crystal lattice is always assumed to be completely rigid, i.e. atomic displacements away from the positions of a perfect lattice are not considered. Now suppose that an atom displaced from its equilibrium site by a small amount. Due to force acting on this atom, it will tend to return to its equilibrium position. This results in **lattice vibrations**. Due to the connections between atoms, the displacement of one or more atoms from their equilibrium positions give rise to a set of correlated vibration waves propagating through the lattice. Consequently, atoms in a crystal lattice can be considered as a series of harmonic oscillators (or pendulums) coupled together by elastic springs. If we displace one pendulum, the springs that couple the pendulums together will cause the neighboring pendelum to move as well and a wave-like motion propagates down the line of oscillators. The frequency of the motion depends on the mass of the pendulums and the strength of the spring constants that connect them. Similar waves pass through a lattice of atoms connected by the binding forces that are responsible for the cohesion of matter. Of course, in this case, the atomic vibrations occur in three dimensions and the whole effect becomes a little hard to visualize. Nevertheless, it is possible to prove that any atomic motion in the crystal can be described by a superposition of waves of different frequencies and wavelengths travelling in different directions. As we know from quantum mechanics the energy levels of the harmonic oscillator are quantized. Similarly the energy levels of lattice vibrations are quantized. The quantum of vibration is called a **phonon**, each having an energy hv, where v is the frequency of the atomic motion. Just as in the pendulum analogy, the frequency of the phonon depends on the wavelength of the distortion, the masses of the atoms, and the stiffness of the "springs" (i.e. binding forces $\propto$ elastic modulus of the material) that connect them.
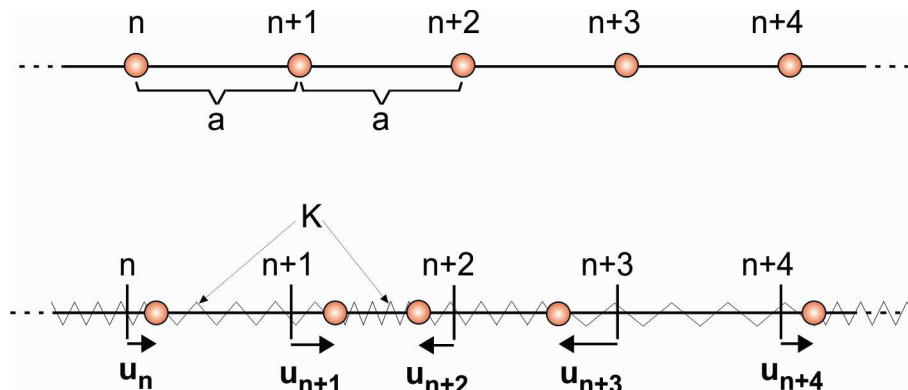

Figure 1: Schematic representation of an infinite monatomic 1D linear spring-chain with lattice constant (average interatomic distance) **a**. Here u_n is the displacement of the n:th atom and **K** is the elastic spring constant.

Mathematically, let us consider an infinite one-dimensional linear chain of atoms of identical mass **M**, connected by springs with equal spring constants **K** as shown in **Fig. 1**. The distance between two atoms in their equilibrium position is **a**, and u_n the displacement of the n:th atom from its equilibrium position. By using the *harmonic approximation* together with *Newton's second law* and making a plane wave ansatz:

$$u_n(t) = Ae^{i(qx - \omega t)}$$

(Eq. 1)

for the solution (**x** = **a·n**), it is possible to deduce (which will be one of your tasks, see below) the

dispersion relation for the phonon frequency as:

$$\omega(q) = 2\sqrt{\frac{K}{M}}\left|\sin\left(\frac{qa}{2}\right)\right| \qquad \textbf{(Eq. 2)}$$

where **q** is the wavevector. For small **q** (i.e. the so-called *Long wavelength limit*, which implies that $\lambda \gg \textbf{a} \Rightarrow \textbf{qa} \ll 1$) the sinus function can be approximated through the argument, resulting in a linear dependence of ω with **q**:

$$\omega(q) = \sqrt{\frac{K}{M}}a|q| \quad \text{valid for} \quad q \ll 1 \qquad \textbf{(Eq. 3)}$$

This dispersion relation (**Eq. 2**) is plotted below in **Fig. 2(a)** and it displays a couple of important properties:

▪ *Reducing to the first Brillouin zone*. The frequency (**Eq. 2**) and the displacement of the atoms do not change when we exchange q by q+2π/a. This means that these solutions are physically identical. This allows us to set the range of independent values of q within the first Brillouin zone, i.e. -π/2 ≤ q ≤ π/2. Within this range of ω(q) is shown in **Fig. 2(a)** where the maximum frequency at the zone boundary is expressed as:

$$\omega_{max}(\pm\pi/2) = \sqrt{\frac{4K}{M}} \qquad \textbf{(Eq. 4)}$$

The frequency is symmetric with respect to the sign change in q, i.e. ω(q) = ω(-q). This is not surprising because a mode with positive q corresponds to the wave traveling in the lattice from the left to the right and a mode with negative q corresponds to the wave traveling from the right tot the left. Since these two directions are equivalent in the lattice the frequency does not change with the sign change in q. At the boundaries of the Brillouin zone q=±π/a the solution represents a standing wave $u_n = (-1)^n e^{-i\omega t}$ where the atoms oscillate in opposite phases depending on whether n is even or odd. The wave moves neither right nor left.

▪ *Phase velocity (v_p) and Group velocity (v_g)* are defined according to:

$$v_p = \frac{\omega}{q} \qquad \text{and} \qquad v_g = \frac{\partial\omega}{\partial q} \qquad \textbf{(Eq. 5)}$$

The physical distinction between the two velocities is that vp is the velocity of the propagation of the plane wave, whereas the vg is the velocity of the propagation of the wave packet. The latter is the velocity for the propagation of energy in the medium. For the particular dispersion relation (**Eq. 2**) the group velocity is given by:

$$v_g = \sqrt{\frac{Ka^2}{M}}\cos\left(\frac{qa}{2}\right) \qquad \textbf{(Eq. 6)}$$

As is seen, the group velocity is zero at the edge of the zone where q=±π/a. Here the wave is standing and therefore the transmission velocity for energy is zero.
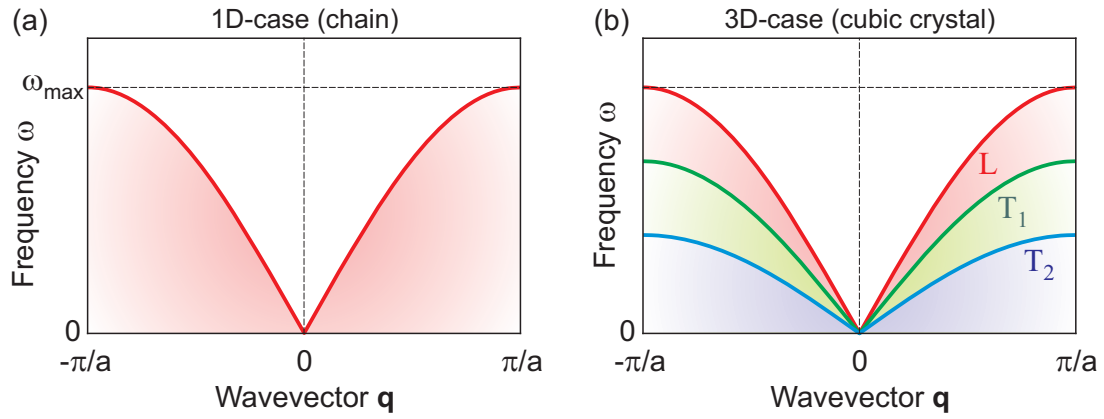
Figure 2: (a) Schematic dispersion curve for a monatomic linear chain with only nearest-neighbor interactions. Note that the frequency is linear for small q and that $\partial\omega/\partial q$ vanishes at the zone-boundaries (i.e. q = $\pm\pi$/a). (b)

These considerations can be extended to the three-dimensional lattice. To avoid mathematical details we shall present only a qualitative discussion. Consider a monatomic Bravais lattice, in which each unit cell has a single atom. The equation of motion of each atom can be written in a manner similar to that of the 1D case. The solution of this equation in three dimensions can be represented in terms of normal modes:

$$\vec{u}_n(t) = \vec{A}e^{i(\vec{q}\vec{r}-\omega t)}$$

(Eq. 7)

where the wave vector **q** specifies both the wavelength and direction of propagation. The vector **A** determines the amplitude as well as the direction of vibration of the atoms. Thus this vector specifies the polarization of the wave, i.e., whether the wave is longitudinal (**A**||**q**) or transverse (**A**⊥**q**). From this we obtain three simultaneous equations involving A_x, A_y and A_z, the components of **A**. These equations are coupled together and are equivalent to a 3×3 matrix equation. The roots of this equation lead to three different dispersion relations, or three dispersion curves, as shown in **Fig. 2(b)**. The three branches differ in their polarization. When **q** points along a high-symmetry direction - for example, the [100], [110] or [111] directions – the situation can once again be considered a 1D chain giving three separate dispersion relations on the same form as **Eq. 2**. The waves may be classified as either pure longitudinal or pure transversal where two of the branches are transverse (**T$_1$** and **T$_2$**) and one is longitudinal (**L**) [again, see **Fig. 2(b)**]. However, along non-symmetry directions the situation is much more complicated and the waves may not be pure longitudinal or pure transversal, but have a mixed character. However, such situation is beyond the scope of this practicum.

Finally, why are phonons important and consequently interesting? The answer to such question is very complex and extensive. However, phonons have a strong impact on several fundamental solid state properties like heat capacity, thermal expansion and transport properties (*e.g.* scattering of electrons or optical absorption). Phonons were also rather famously found responsible for acting as the 'glue' between electrons in the formation of Cooper pairs in conventional superconductors.

5. Inelastic Neutron Scattering (INS)

One of the great advantages of neutrons as a probe in condensed matter physics is that they can be used to measure details of atomic and molecular motions by using so-called inelastic neutron scattering (**INS**), in which the neutron exchanges energy with the atoms in a material. When a neutron is scattered by a crystalline solid, it can absorb or emit an amount of energy equal to a quantum of phonon energy, hv. This gives rise to inelastic coherent scattering in which the neutron energy before and after the scattering differ by an amount, hv (i.e. **energy transfer**). In most solids v is a few terahertz (THz), corresponding to phonon energies of a few meV (1 THz corresponds to an energy of 4.18 meV). Because neutrons used for INS also have energies in the meV range, scattering by a phonon causes an appreciable fractional change in the neutron energy, allowing accurate measurement of phonon frequencies. For INS, the neutron has different velocities, and thus different wavevectors, before and after it interacts with the sample. To determine the phonon energy and the scattering vector, **Q**, we need to determine the neutron wavevector before (**k_i**, i = *initial*) and after (**k_f**, f = *final*) the scattering event. Several different methods are available but the workhorse instruments for this type of measurement are called triple-axis spectrometers.
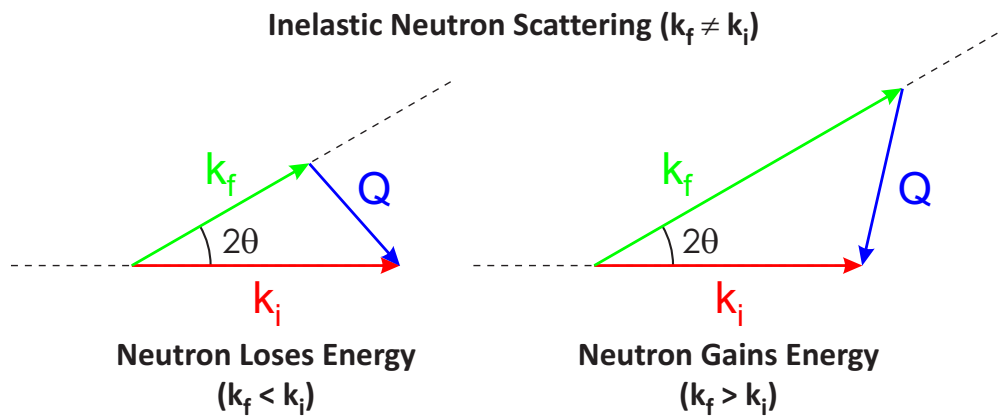
Inelastic Neutron Scattering ($k_f \neq k_i$)

Neutron Loses Energy
($k_f < k_i$)

Neutron Gains Energy
($k_f > k_i$)

Figure 3: Scattering triangles for inelastic neutron scattering (INS) in which the neutron either loses energy (**k_f** < **k_i**) or gains energy (**k_f** > **k_i**) during the interaction with the sample. In both elastic and inelastic scattering events, the neutron is scattered through the angle 2θ, and the scattering vector is given by the vector relationship **Q** = **k_i** − **k_f**.

The three-axis spectrometer (**TAS**) can be considered the mother of all crystal spectrometers and they are the most versatile instruments for performing INS experiment. They allow for a controlled access to both the momentum **Q** = **k_i**−**k_f** as well as energy transfer $\hbar\omega$ = E_i−E_f. During an TAS experiment (see **Fig. 4**) an incident neutron beam of well defined wavevector **k_i** is selected from the white spectrum of the neutron source by the monochromator crystal (first axis). The monochromatic beam is then scattered from the crystalline sample (second axis). The intensity of the scattered beam with wavevector kf is reflected by the analyzer crystal (third axis) onto the neutron detector (^{3}He-tube), thereby defining the energy transfer $\hbar\omega$ as well.
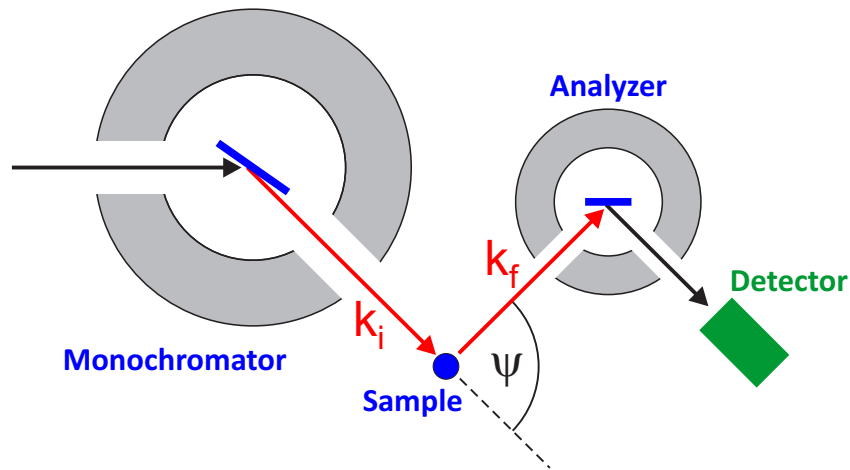
Triple-axis Spectrometer (TAS)

Figure 4: Schematic layout of a neutron triple-axis spectrometer.

As already mentioned, the main advantage of a triple-axis spectrometer is that experimental data can be acquired at any pre-determined point in the reciprocal space (Q) for a selected energy transfer $\hbar\omega$. Practically, data is recorded by scanning one or both of the variables along a chosen direction. One usually chooses between ⬜ Constant-**Q** scans where $\hbar\omega$ is scanned while keeping **Q** fixed or ⬜ Constant-energy scans where $\hbar\omega$ is kept at a fixed value and Q is scanned along a selected direction in reciprocal space (see **Fig. 5**). By performing one or both these types of scans the dispersion relation $\hbar\omega(Q)$ for a single crystal sample can be extracted in a very controlled manner. Of course also more complicated scans are possible where **Q** and $\hbar\omega$ are scanned simultaneously. Since on a TAS all quantities ($\mathbf{k_i}$, $\mathbf{k_f}$, ψ) that defines the momentum and energy transfers are in principle variable, there are many different ways to measure the intensity at the same selected [**Q**,$\hbar\omega$] point. The most common method, and also the one we will use in this practicum, is however to keep $\mathbf{k_f}$ fixed.
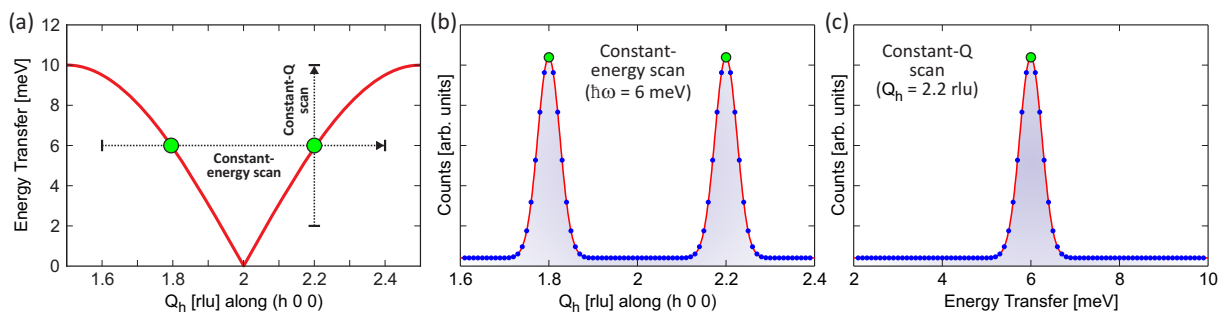

Figure 5: (a) Schematic view of how two points of the phonon dispersion curve can be measured using either (b) constant-energy scan or (c) constant-**Q** scan. By performing multiple scans it is possible to map out the complete dispersion (see below).

6. Description of the Practicum

This advanced practicum will be performed using either the **TASP** or **RITA-II** cold neutron triple-axis spectrometer at the Swiss Spallation Neutron Source (SINQ). The main topic is to measure the phonon dispersion curve of a lead (Pb) crystal using inelastic neutron scattering (INS). The practicum will be performed with help and under the supervision of your local contact. All the necessary information regarding how to technically control the neutron spectrometer will be given to you on site at the day of the practicum. During and after the practicum there are a series of different tasks to perform, which should also be a part of your final report. Please read through the list of specific tasks for your report (next chapter) before starting the experimental work.

Your first practical task will be to set up the spectrometer and to verify the alignment of the instrument by performing an energy (constant-q) scan around the elastic line of a Vanadium calibration sample. Once the instrument is in order your will align the Pb crystal (see photo to the right) by finding several nuclear Bragg reflections using the triple-axis neutron spectrometer. Nominal values for the lattice parameters are given above but you should experimentally refine such values using the spectrometer.

When the Pb crystal is aligned, you will acquire a series of either constant-q or constant-energy scans (or a combination) in order to map out the phonon-dispersion ($h\nu$ as a function of q) along a selected high-symmetry direction ($h\ 0\ 0$). How to perform a scan will be shown to you by the local contact. However, just as an example, the two scans shown above in **Fig. X** are at the TASP instrument performed by running the following two text commands in the SIX-prompt:

Constant-energy scan (at $\hbar\omega = 6$ meV): `sc qh 2 0 0 6 dqh 0.01 0 0 0 np 81 mn 5000`

Constant-Q scan (at $Q_h = 2.2$): `sc qh 2.2 0 0 6 dqh 0 0 0 0.1 np 81 mn 5000`

Data files resulting from such scans are stored onto a PSI-server using a simple ASCII format and in principle it is possible to use almost any computer program to analyze the data (e.g. Matlab, Origin, Igor etc.). For the purpose of this practicum it is, however, recommended that you use the local program that is simply called **FIT**. This is installed on the computer that you will use to perform the actual scans and it is very convenient to use for performing quick and basic fits to the data (see screen-shot in **Fig. 6**). The program is very easy to learn and your local contact will show you how. If you wish to prepare in advance there is an online documentation that you find here:

It is highly recommended that the students perform most of the data-analysis (at least fitting of the raw-data) during the practicum since then your local contact can assist you. However, if for some reason you need to access the data-files after the practicum this is certainly possible. The simplest way is to remotely access the data-server using either ssh (ssh client on a Windows machine e.g. **Bitvise**) using the following:

Host: **sinquser-sl6.psi.ch**   Login: **sinquser**   Pass: **sinquser**

Then you will need to change to the folder: **/afs/psi.ch/project/sinqdata/xxxx/yyyy/**

where **xxxx** is the year of interest (e.g. **2013**) and **yyyy** the instrument (e.g. **tasp**). Then simply download the datafiles that you need.
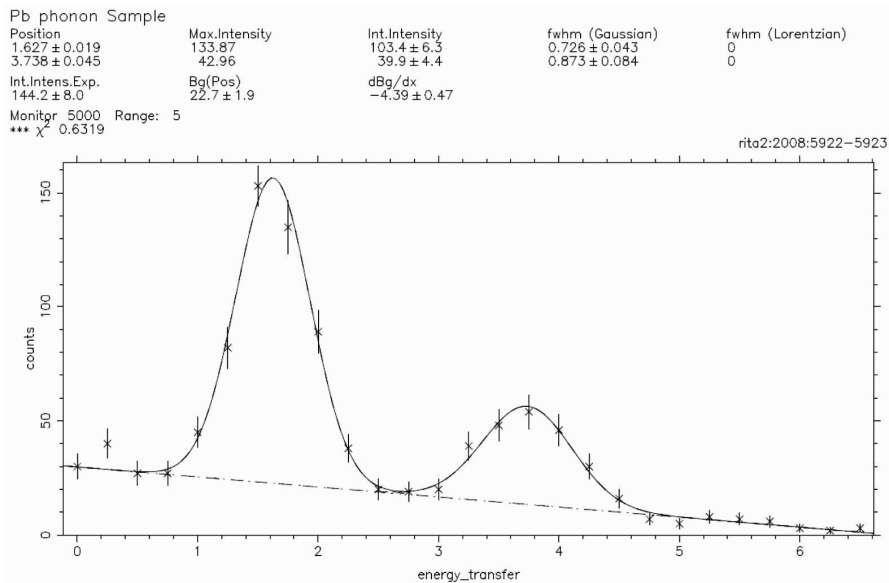
Figure 6: Screenshot from the FIT program showing how a constant-q scan is fitted by two Gaussian functions. The positions (energy transfer) showed in the top left corner together with the **q**-point at which the scan was performed will yield two unique (E,q) points of the phonon dispersion curves [see **Fig. 7(a)** below]. Remember to also record the error-bars of the position!

In your data you will observe 1-4 peaks (depending on which scan you decide to perform). Such peaks are usually fitted using a simple Gaussian function with a sloped background signal (see **Fig. 6**). From the fits acquired using the FIT program you will have a series of **E** vs. **q** datapoints. The dispersion curve is obtained by simply plotting such points in an **E** vs. **q** diagram [see **Fig. 7(a)**]. However, having the rest of your practicum tasks in mind, a useful way to transform your data is shown in **Fig. 7(b)**.
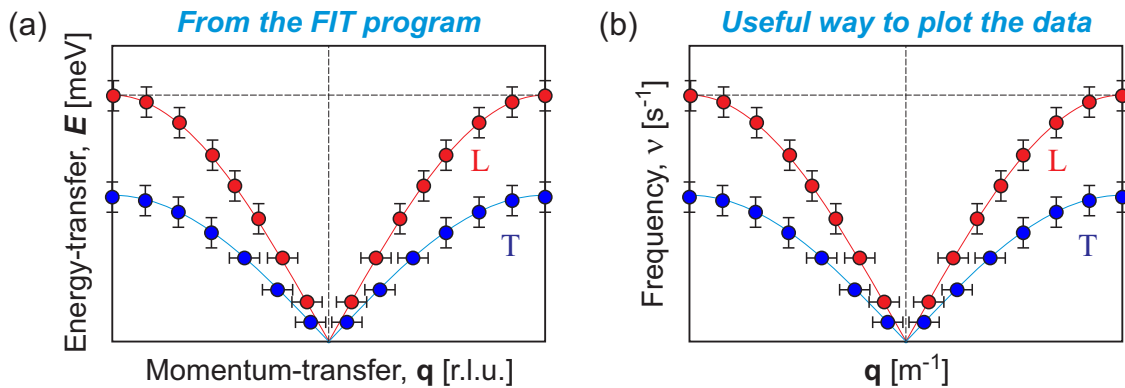

Figure 7: (a) Schematic view of how the extracted points obtained from fits of the data are plotted as phonon dispersion. (b) Useful way to plot the data from the viewpoint of extracting further properties in a straightforward manner (units!).

11

7. Specific Tasks for the Report

The tasks below should be completed during/after you practicum. The experimental procedure, theoretical deductions as well as results from experiment and data analysis should be presented in a written report. Below are some specific points that should be addressed in the report.

- Your report should partly be a logbook of your conducted experiment and should contain a description of how you have performed the measurement. This includes a summary of instrumental settings (collimators, slits, distances, neutron energy) that were used as well as a complete list of file numbers with a one-line description of what scan that file represent. Further, any unexpected issues e.g. beam-dump, instrument problems or realignment of sample should be noted. This is how a scientific experiment is normally performed and the current practicum will be conducted using the same approach.

- Comment on why we aligned the Pb-crystal using the (200) and not *e.g.* the (100) reflections. Your report should also contain the experimentally determined lattice parameter.

- Draw a cut (plane) through the first Brillouin zone of Pb that corresponds to the scattering plane used in your neutron experiment.

- In this Brillouin zone plane, plot the **q**-points for which you have acquired your neutron data. Note that if you did not acquire your data from the first zone; simply extend the plot from above so as to also include second, third…

- Deduce **Eq. 2** starting from a single harmonic oscillator, Newton's second law of motion and the solution ansatz of **Eq. 1**. Specifically explain how the issue of boundary conditions is commonly dealt with.

- From each of the neutron spectra, fit the peaks and extract the position ($\hbar\omega$,**q**) of the different phonon modes. A plot of each scan including the fit results should be presented in your report.

- Plot your measured phonon dispersions (**including error-bars!!!**). Can you comment on why there is only one transversal (T) phonon-mode and not two?

- Determine the speed of sound in Pb (transversal as well as longitudinal) and compare your results to values found in the literature.

- Determine as many of the elastic stiffness constants (c_{11}, c_{12}, c_{44}) as you possibly can from the measured phonon dispersion and compare your results to values found in the literature *e.g.* ultrasonic measurements. If you cannot determine some of the constants, explain why! **(Hint:** Kittel **[1]** is a good start)