PUMA

Thermal Triple Axis Spectrometer

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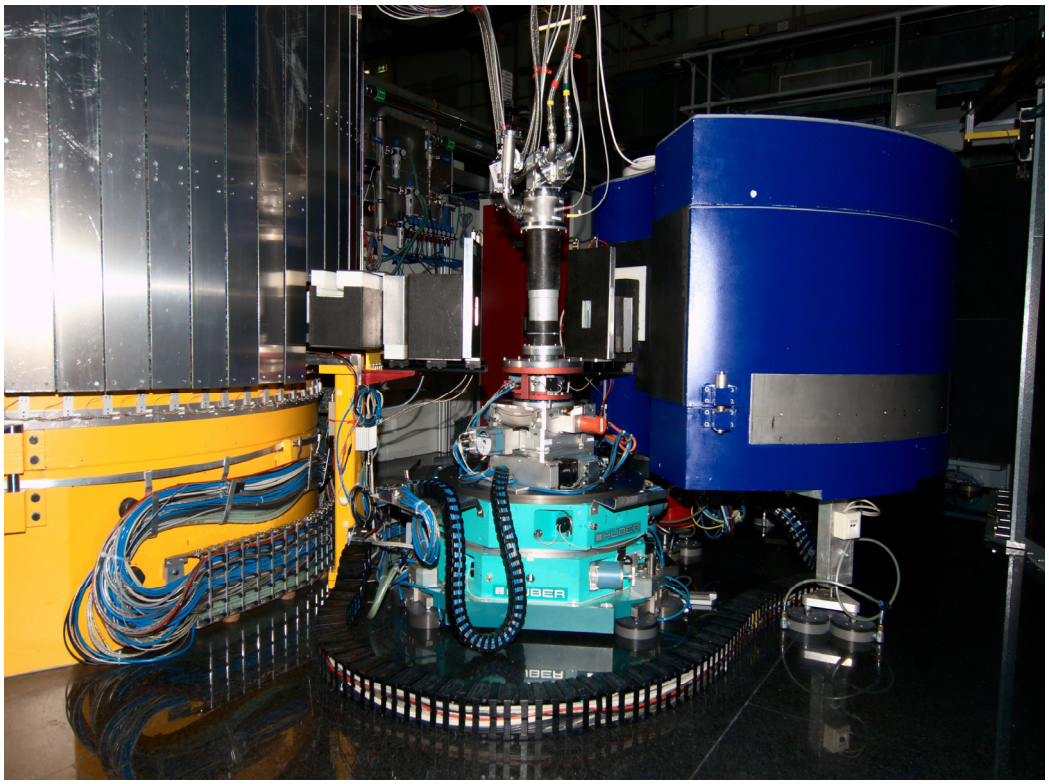


Manual of the JCNS Laboratory Course Neutron Scattering

Contents

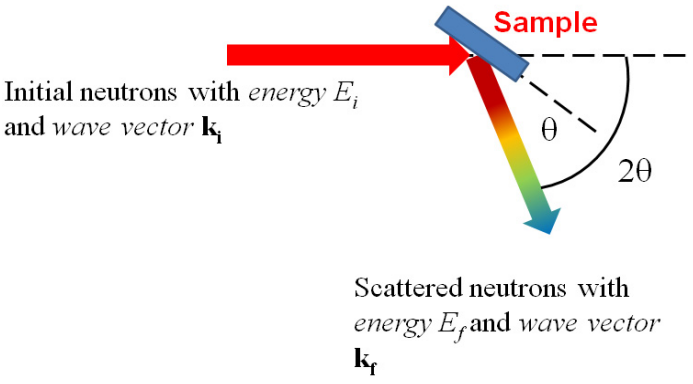
1. Introduction

Excitations in crystals can be described using formalism of dispersion relations of the normal modes or quasi-particles (phonons, magnons, etc.). These relations contain the most detailed information on the intermolecular interactions in solids.

The result of a neutron scattering experiment is the distribution of neutrons that have undergone an energy exchange $\hbar\omega = E_i - E_f$, and a wave vector transfer, $\mathbf{Q} = \mathbf{k_i} - \mathbf{k_f}$, after scattering by the sample.:

$$\frac{d^2\sigma}{d\Omega d\omega}(2\theta,\omega) = N\frac{k_f}{k_i}\left\{\frac{\sigma_{coh}}{4\pi}S_{coh}(\mathbf{Q},\omega)+\frac{\sigma_{inc}}{4\pi}S_{inc}(\mathbf{Q},\omega)\right\} \quad (1)$$

σ_{coh} is coherent scattering cross section, σ_{inc} is incoherent scattering cross section. They are constants that can be found in tables (http://www.ncnr.nist.gov/resources/n-lengths/). $S(\mathbf{Q},\omega)$ functions depend only on the structure and dynamics of the sample and do not depend on the interaction between neutrons and the sample. $S_{inc}(\mathbf{Q},\omega)$ reflects individual motions of atoms. $S_{coh}(\mathbf{Q},\omega)$ provides the information on the structure and collective excitations in the sample.



Energy transfer

$\hbar\omega = E_i - E_f$

Momentum transfer

$\mathbf{Q} = \mathbf{k_0} - \mathbf{k_f}$

$$k = \frac{\sqrt{2mE}}{\hbar} = \frac{2\pi}{\lambda}$$

$$Q = \sqrt{k_i^2 + k_f^2 - 2k_i k_f \cos 2\theta}$$

If $k_i = k_f$

$$Q = 2k_i \sin\theta = \frac{4\pi}{\lambda}\sin\theta$$

The triple axis spectrometer is designed for measuring the $S_{coh}(\mathbf{Q},\omega)$ in monocrystals. Therefore this function is of special interest for us.

2. Elastic scattering and Structure of Crystals

In the case of *elastic scattering,* when $\omega = 0$ ($k_i = k_f$) only neutrons, that fulfil the Brags law are scattered by the sample:

$$n\lambda = 2d_{hkl}\sin\theta_{hkl}, \tag{2}$$

where λ is a wavelength of neutron, d_{hkl} is a distance between crystal planes described by corresponding Miller indexes *hkl*. θ_{hkl} denotes the angle between incoming (outgoing) scattering beam and the (hkl) plane.

For the analysis of the scattering processes in crystals it is convenient to use the concept of the *reciprocal space*. For an infinite three dimensional lattice, defined by its primitive vectors $\boldsymbol{a_1}$, $\boldsymbol{a_2}$ and $\boldsymbol{a_3}$, its reciprocal lattice can be determined by generating three reciprocal primitive vectors, through the formulae:

$$\begin{aligned}
\mathbf{g}_1 &= 2\pi \frac{\mathbf{a}_2 \times \mathbf{a}_3}{\mathbf{a}_1 \cdot \mathbf{a}_2 \times \mathbf{a}_3} \\
\mathbf{g}_2 &= 2\pi \frac{\mathbf{a}_1 \times \mathbf{a}_3}{\mathbf{a}_2 \cdot \mathbf{a}_1 \times \mathbf{a}_3} \\
\mathbf{g}_3 &= 2\pi \frac{\mathbf{a}_1 \times \mathbf{a}_2}{\mathbf{a}_3 \cdot \mathbf{a}_1 \times \mathbf{a}_2}
\end{aligned} \tag{3}$$

Note the denominator is the scalar triple product. Geometrically, the scalar triple product $\mathbf{a_1(a_2 \times a_3)}$ is the volume of the parallelepiped defined by the three vectors.

Let us imagine the lattice of points given by the vectors $\mathbf{g_1}$, $\mathbf{g_2}$ and $\mathbf{g_3}$ such that $\boldsymbol{\tau}$ is an arbitrary linear combination of these vectors:

$$\boldsymbol{\tau} = h\mathbf{g_1} + k\mathbf{g_2} + l\mathbf{g_3}\,, \tag{4}$$

where *h,k,l* are integers. Every point of the reciprocal lattice, characterized by $\boldsymbol{\tau}$ corresponds in the position space to the equidistant set of planes with Miller indices (*h,k,l*) perpendicular to the vector $\boldsymbol{\tau}$. These planes are separated by the distance

$$d_{hkl} = \frac{2\pi}{\left|\boldsymbol{\tau}_{hkl}\right|} \tag{6}$$

The Brag's condition for diffraction can be expressed in the following vector form:

$$\mathbf{Q} = \boldsymbol{\tau}_{hkl} \tag{7}$$

A useful construction for work with wave vectors in reciprocal space is the Brillouin zone (BZ). The BZ is the smallest unit in reciprocal space over which physical quantities such as phonon or electron dispersions repeat themselves. It is constructed by drawing vectors from one reciprocal lattice points to another and then constructing lines perpendicular to these vectors at the midpoints. The smallest enclosed volume is the BZ.

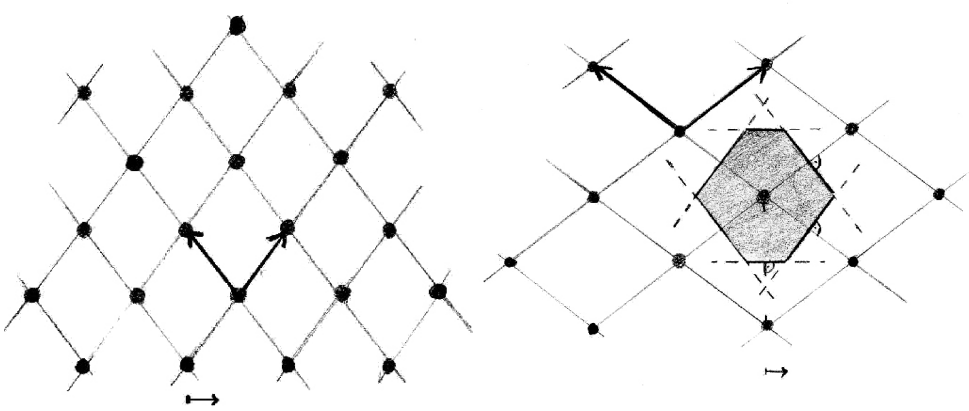


Fig.1 Real (left) and reciprocal (right) two dimensional lattices and BZ (gray area)

3. Inelastic Neutron Scattering and Phonons

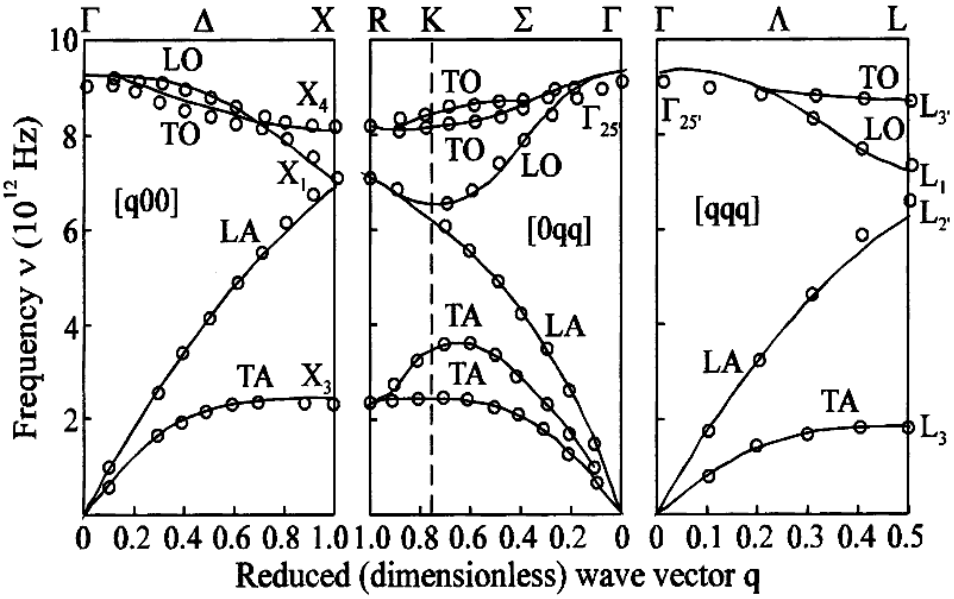


Fig.2 Phonon dispersion curves for Ge.

Atomic vibrations in a crystal can be analysed in terms of lattice waves which are the normal modes of the crystal. The frequencies of normal modes ω are related to their wave vectors $\mathbf{q}$ ($q = 2\pi/\lambda$) by the dispersion relations

$$\omega = \omega_j(\mathbf{q}), \tag{7}$$

where the index j denotes a particular branch. For a crystal with N atoms per primitive unit cell there are $3N$ branches of the frequency spectrum. Three branches are acoustic ones for which $\omega \to 0$ as $\mathbf{q} \to 0$; the other $3N$-3 are branches are optical branches for which ω tends to a finite value as $\mathbf{q} \to 0$. In certain directions of high symmetry the normal vibrations are strictly transverse or longitudinal. The energy quantum $\hbar\omega$ is called *phonon* in analogy to the phonon for electromagnetic waves.

If we want to measure the frequency of a phonon ω for a certain $\mathbf{q}$, the basic scattering conditions must fulfil the energy and momentum conservation laws:

$$E_i - E_f = \frac{\hbar}{2m_n}(k_i^2 - k_f^2) = \pm\hbar\omega(\mathbf{q}) \qquad (9)$$

$$\mathbf{Q} = \mathbf{k_i} - \mathbf{k_f} = \mathbf{G} \pm \mathbf{q}$$

When the above conditions are fulfilled, the function $S_{coh}(\mathbf{Q},\omega)$ shows a peak. We can held **Q** constant and vary $\mathbf{k_i}$ ($\mathbf{k_f}$) to measure intensity of scattered neutrons at different energy transfers. In order to keep **Q**, and thus **q**, constant while varying $\mathbf{k_i}$, the scattering angle must change as well as the relative orientation of the crystal with respect to $\mathbf{k_f}$.

The intensity of neutrons scattered by phonon is proportional to the square of the dynamical structure factor $F(\mathbf{Q})$:

$$S_{coh}(\mathbf{Q},\omega) \sim |F(\mathbf{Q})|^2 = \left| \sum_\kappa b_\kappa \frac{\mathbf{Q}\cdot\mathbf{e}_\kappa(\mathbf{q}_j)}{\sqrt{m_\kappa}} \exp(-W_k)\exp(-i\mathbf{q}\mathbf{r}_\kappa) \right|^2, \qquad (10)$$

Where sum is taken over all atoms in unit cell with coordinates $\mathbf{r}_k$, exp(-W) is a Debye-Waller factor, $\mathbf{e}_k$ denotes the polarization vector of the phonon. The scalar product $\mathbf{Q}\cdot\mathbf{e}_\kappa(\mathbf{q}_j)$ means that only lattice vibrations polarized along the momentum transfer are visible. This makes possible to distinguish transverse (TA) and longitudinal (LA) acoustic modes. For TA modes $\mathbf{e}\perp\mathbf{q}$, and therefore **Q** must be perpendicular to **q**, while for a LA mode, one must take $\mathbf{Q} \,||\, \mathbf{q}$ (Fig. 3)

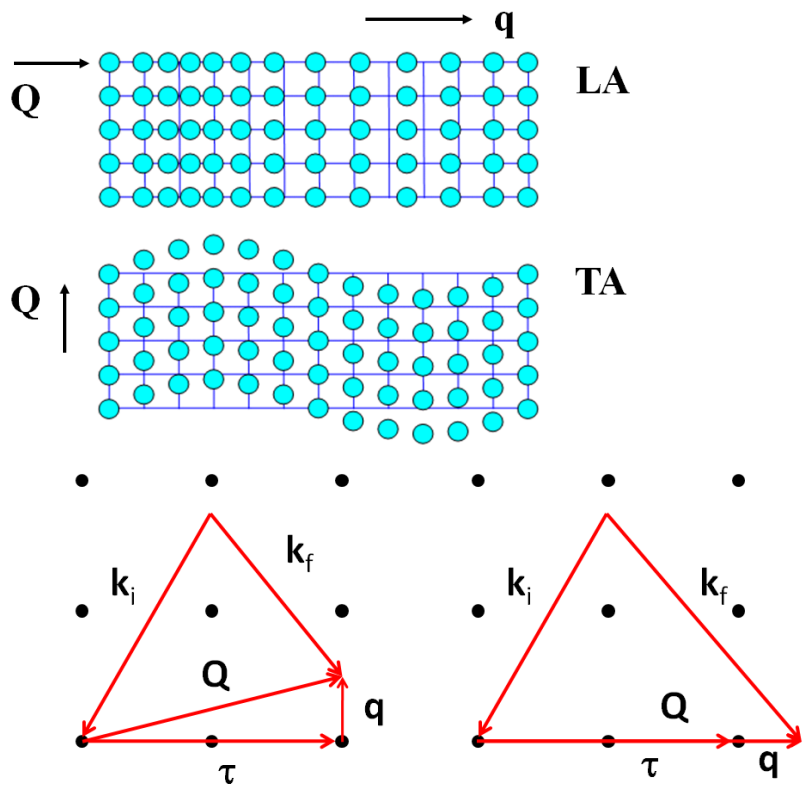


Fig. 3 Top: LA and TA phonons. Bottom: Neutron scattering diagram in the reciprocal space for TA (left) and LA phonons

4. Triple Axis Spectrometer PUMA

The three-axis instrument is the most versatile instrument for use in inelastic scattering because it allows one to probe nearly any coordinates in energy and momentum space in a precisely controlled manner. The three axes correspond to the axes of rotation of the *monochromator* (axis1), *the sample* (axis2), and *the analyzer* (axis3). The monochromator crystal selects neutrons with a certain energy from the white neutron beam emanating from the reactor. The monochromatic beam is then scattered off from the sample (second axis). The neutrons scattered by the sample can have a different energy from those incident on the sample. The energy of these scattered neutrons is then determined by the analyzer crystal (third axis). All three angles (θ_M, θ_S, θ_A) can vary during an experiment, the sample table and analyzer are equipped with air pads, so that they can glide over the "Tanzboden" (dancing floor). Below, we describe in detail each component of a triple-axis spectrometer.

Monochromator

A crystal monochromator is used to select neutrons with a specific wavelength. Neutrons with this wavelength interact with the sample and are scattered off at a similar (elastic) or different wavelength (inelastic). The energy of the neutrons both incident on and scattered from the sample is determined by Bragg reflection from the monochromator and analyzer crystals, respectively. For a specific Bragg plane (hkl) characterized by an interplanar spacing d_{hkl}, the crystal is rotated about a vertical axis. A pyrolytic graphite with d_{002} = 3.35 Å (PG(002)) and a copper with d_{220} = 1.28 Å (Cu(220)) monochromators are available at PUMA. The angular range of the monochromator $2\theta_M$ is of 15° - 115°. The PG(002) is usually used for energies below 50meV (λ>1.3Å). For higher incident energies the Cu(220) can be used.

Sample table

The sample table from the company Huber provides a possibility to vary independently both $2\theta_S$ and θ_S. It is equipped with a goniometer moving the sample in the three translation axes *x*, *y* and *z* and tilting. The tilt angle is ±15°. Single crystal experiments can be performed with an Euler cradle at PUMA. The sample environment includes magnets, pressure cells, cryostats and high temperature furnace.

Analyzer

Like the monochromator, the PG(002) analyzer consist of 20x5 separate analyzer crystal plates are mounted in an aluminum frame. There is an option to measure with the flat or horisontaly and verticaly focused analyser. The angular range of the analysator $2\theta_M$ is of -130° - 130°.

Detector and monitor

The detector consists of five counter tubes which are filled with a ^{3}He pressure of 5 bar. To be able to monitor the neutron flux incident on the sample, a low-efficiency neutron counter monitor is usually placed before the sample. Such a monitor is required so that flux variation caused by, for example, the reactor power fluctuations and the change in reflectivity of the monochromator with neutron wavelength can be automatically corrected for.

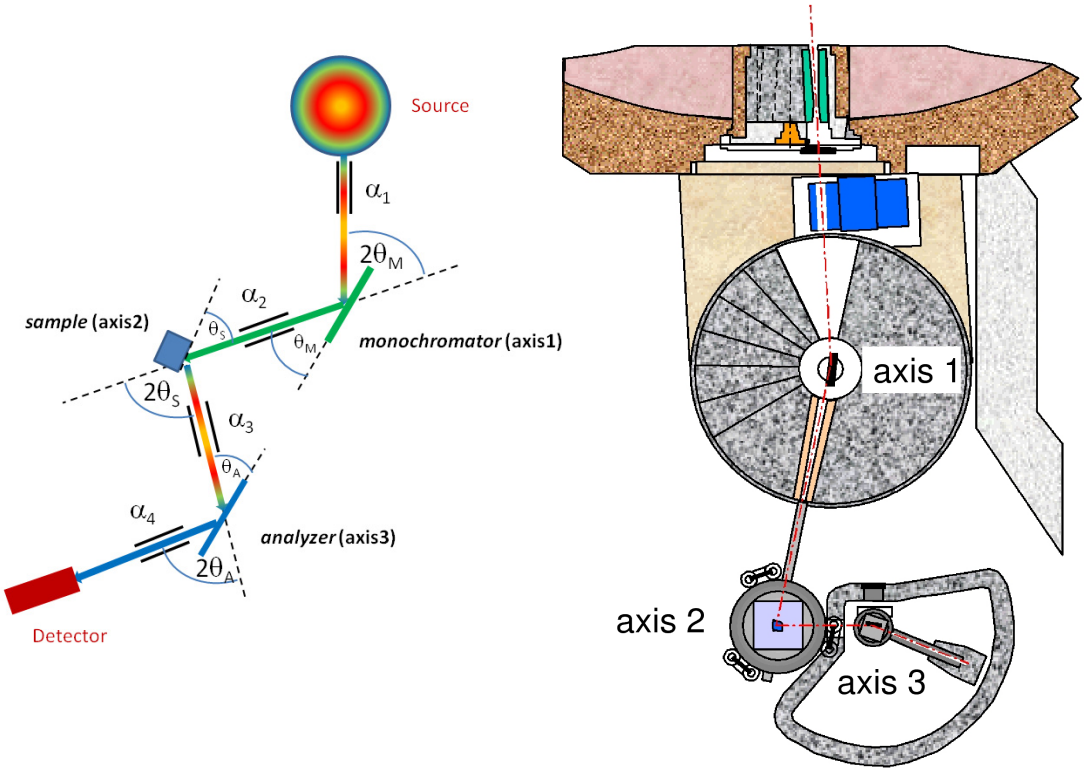


Fig.4 PUMA spectrometer.

Slits, Collimators, Filter

Additional components like slits or collimators are used to define the beam cross section. Collimators (α1- α4) are used for the improvement of the resolution and to specify the beam divergence. They consist of multiple parallel arranged Gd_2O_3 coated foils with a defined angle to the beam. The angular divergence of the collimator in the horizontal plane α is defined by the distance between foils Δd and the length of the collimator l (tan $\alpha = \Delta d / l$). Different collimators with a horizontal divergence between 10' and 60' are available at the instrument.

One of the problems of the TAS method is the possible presence of higher harmonics in the neutron beam. Higher harmonics arise from higher order (hkl) in Bragg's law (2). This means that if the monochromator (analyzer) crystal is set to reflect neutrons with a wavelength of λ from a given (hkl) plane, it will also reflect neutrons with wavelength λ/n. This leads to the appearance of several types of spurious peaks in the observed signal. Different filters are used to eliminate the high-order neutrons and to reduce the background. There are a sapphire filter (Al_2O_3) and an erbium filter (Er) at PUMA. They are installed in front of the monochromator. Sapphire filter is used wavelengths $\lambda > 1$ Å and reduce the background inducing by the epithermal neutrons. Erbium filter is suitable as $\lambda/2$ filter for λ between 0.5 and 1Å as well as $\lambda/3$ filter for λ between 0.7 and 1.6Å.

Components	Axis	PUMAs notation	Description
Monochromator M	θ_M	mth	Monochromator Theta
	$2\theta_M$	mtt	Monochromator 2Theta
		mtx, mty	Monochromator Translation x-, y- direction
		mgx, mgy	Monochromator Goniometer x-, y- direction
		mfh, mfv	Monochromator Focus horizontal, vertical
Sample S	θ_S	psi	Sample Theta
	$2\theta_S$	phi	Sample 2Theta
		stx, sty, stz	Sample Translation x-, y-, z- direction
		sgx, sgy	Sample Goniometer x-, y- direction
Analyzer A	θ_A	ath	Analyzer Theta
	$2\theta_A$	att	Analyzer 2Theta
		atx, aty	Analyzer Translation x-, y- direction
		agx, agy	Analyzer Goniometer x-, y- direction
		afh	Analyzer Focus horizontal
Collimators		alpha1 – alpha4	Collimation

5. Experiment Procedure

The aim of the experiment is to measure acoustic phonons in a germanium sample. The phonons will be measured for [110] (LA) and [001] (TA) directions in [220] BZ.
The experimental procedure shall contain the following steps:

Sample alignment

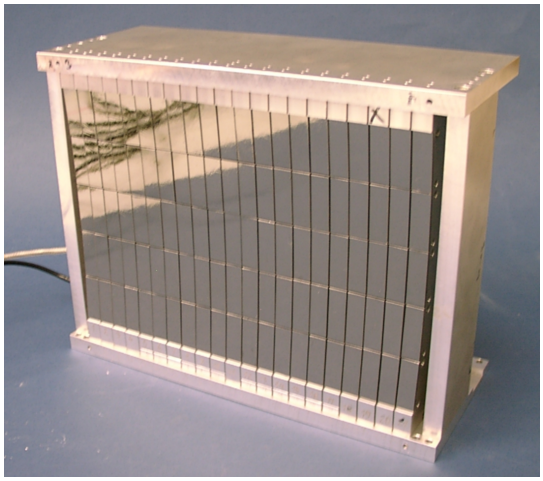
It is very difficult to align a sample with triple axis spectrometer, if the sample orientation is absolutely unknown. A sample must be pre-aligned, this means that the vertical axis of the sample must be known and roughly perpendicular to the 'Tanzboden'. Than we shall do the following steps:
- Inform the control program of the spectrometer about a scattering plane of the sample. One must set two reciprocal vectors (in our case [110] and [001]) laying in the scattering plane.
- Drive spectrometer (θ_M, $2\theta_M$, θ_S, $2\theta_S$, θ_A, $2\theta_A$,) to the position corresponding to [220] reflection.
- Scan θ_S and find the Brag's peak.
- Scan corresponding goniometer axes to maximize intensity of the peak.
- Do the same for other reflection [004].
- Change the offset of the θ_S so that the nominal θ_S values correspond to intensity maxima for the above reflections.

Phonons measurements

For our measurements we will chose the const-k_f configuration with $k_f = 2.662$ Å^{-1} ($E_f = 14.68$ meV). This means that we will scan the energy transfer $\hbar\omega = E_i - E_f$ by varying incident energy E_i (k_i). We are going to use PG(002) monochromator.

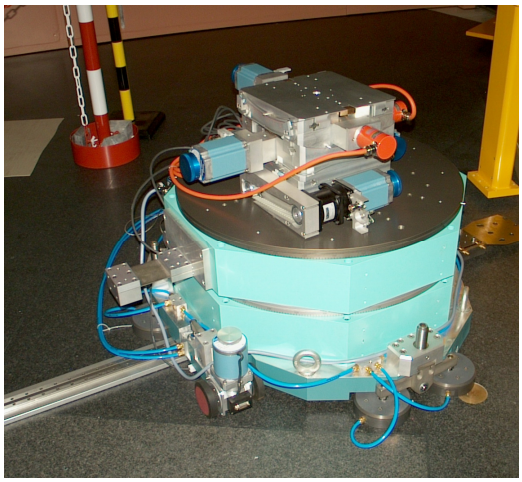
For LA phonon we will do constant-**Q** scans in the energy transfer range $\hbar\omega$ = 0 – 21 meV (0 – 8 THz) for the following points:
Q(r.l.u.) = (2.1, 2.1, 0), (2.2, 2.2, 0), (2.3, 2.3, 0), (2.4, 2.4, 0), (2.5, 2.5, 0), (2.6, 2.6, 0), (2.7, 2.7, 0), (2.75, 2.75, 0).

For TA phonon we will do constant-**Q** scans in the energy transfer range $\hbar\omega$ = 0 – 15 meV (0 – 3.6 THz) for the following points:
Q(r.l.u.) = (2, 2, 0.2), (2, 2, 0.3), (2, 2, 0.4), (2, 2, 0.5), (2, 2, 0.7), (2, 2, 0.8), (2, 2, 0.9), (2, 2, 1).

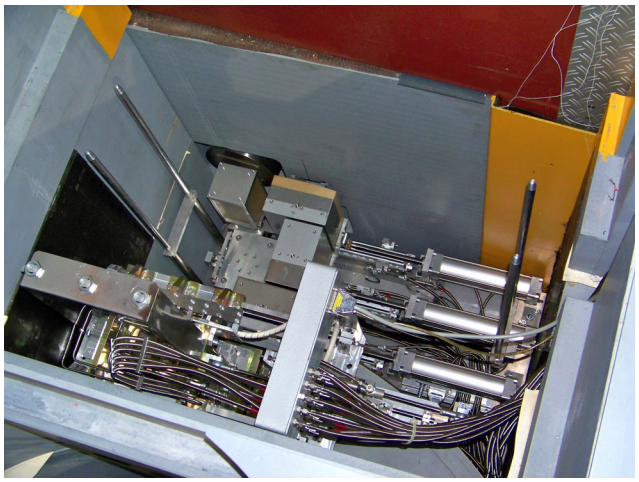


a) PG Analyzer

b) Soller collimator



c) Sample table



d) Shutter, filters and collimators

e) Analyzer and Detector

f) Detector, consists of 5 ^{3}He tubes

Fig 5 Elements of PUMA

6. Preparatory Exercises

1. Calculate angles θ_M, $2\theta_M$, θ_S, $2\theta_S$, θ_A, $2\theta_A$ for the reflections [220] and [002] of *germanium* (cubic-diamond, a = 5.66 Å), supposing that k_f = 2.662 Å^{-1} = const, minichromator is PG(002).

2. Before doing a scan it is important to check that all point in **Q** - $\hbar\omega$ space are available, instrument angles do not exceed high or low limits. Also, an experimental scientist must be sure that the moving instrument will not hit walls or any equipment. Calculate instrument parameters (θ_M, $2\theta_M$, θ_S, $2\theta_S$, θ_A, $2\theta_A$) for the momentum transfers **Q** (r.l.u.) = (2.1, 2.1, 0), (2.75, 2.75, 0) and energy transfers $\hbar\omega$ = 0 and 21 meV. This can be done using an online triple-axis simulator:
http://www.ill.eu/instruments-support/computing-for-science/cs-software/all-software/vtas/

7. Experiment-Related Exercises

1. Plot obtained spectra for each **Q** as a function of energy (THz). Fit the spectra with Gaussian function and find centers of the phopon peaks. The obtained phonon energies plot as a function of **q.**
2. Why triple-axis spectrometer is the best instrument to study excitations in single crystals?
3. During this practicum we do not consider some problems that are very important for planning experiments with a triple axis instrument such as *resolution* and *intensity zones* [2]. Persons who have a strong interest to the triple-axis spectroscopy should study these topics by oneself. Advanced students should be able to explain our choice of Brillouin zone and parameters of scans for the phonon measurements.

Useful formula and conversions

1 THz = 4.1.4 meV

$n\lambda = 2d_{hkl}\sin\theta_{hkl}$,

$$d_{hkl} = \frac{2\pi}{|\boldsymbol{\tau}_{hkl}|}$$

$$\mathbf{Q} = \mathbf{k_0} - \mathbf{k_f}$$

$$Q = \sqrt{k_i^2 + k_f^2 - 2k_i k_f \cos 2\theta}$$

If $k_i = k_f$ (elastic scattering) $Q = 2k_i \sin\theta = \frac{4\pi}{\lambda}\sin\theta$

E [meV] = 2.072 k^2 [Å^{-1}]

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