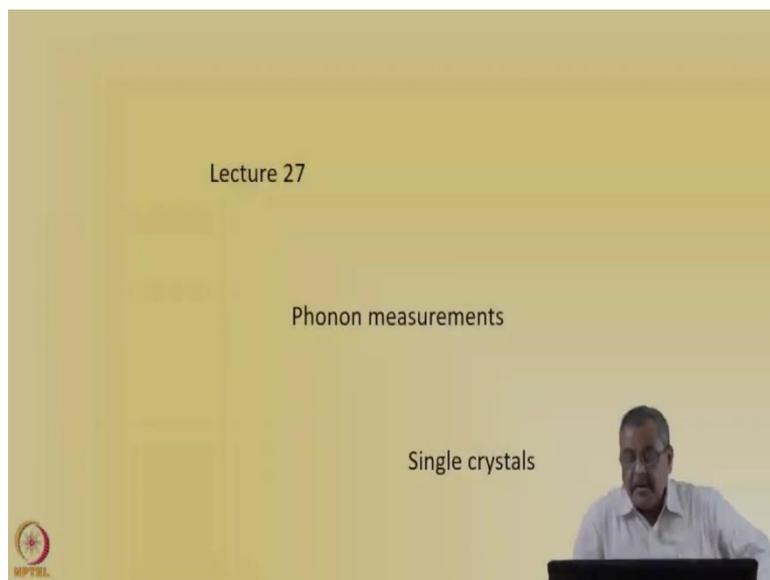**Neutron Scattering for Condensed Matter Studies**
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Week 11: Lecture 27A

Keywords: Triple-axis spectrometer, Mosaic crystal, Single crystal

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In this lecture, we will continue with phonon measurements and we will be discussing single crystals. Primarily, I will be talking about doing experimental measurements of phonon dispersion curves using neutrons.

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Triple Axis Spectrometer
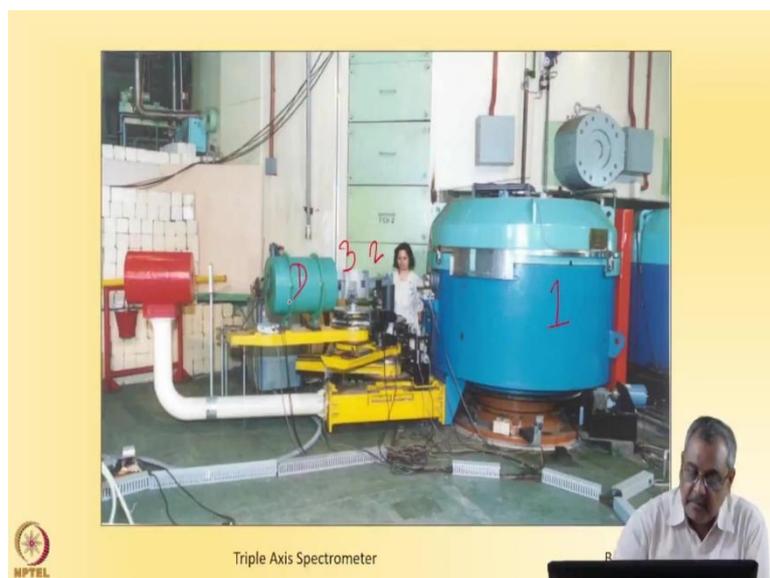
It is important that first I discuss with you the way the experiments are being done, I have told you several times and showed you these spectrometers known as triple axis spectrometers. It was designed by B. N. Brockhaus for studying inelastic dynamics or inelastic neutron scattering to reveal dynamics in various materials or in condensed matter in general. As I told you that monochromator drum is the first axis, the sample is the second axis and then the analyzer around the sample, which finds out the final energy is the third axis in a triple axis spectrometer and then there is a detector, working in $\theta - 2\theta$ mode around the analyzer crystal.

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Triple Axis Spectrometer

Mosaic Crystals

$[1,1,1] \rightarrow \lambda$

Cu monochromator

$[2 2 2] \quad \frac{\lambda}{2}$

$2d \sin\theta = n\lambda$

Improved intensity
Compromised resolution
$\lambda \pm \Delta\lambda$

$N(\lambda)$

$\lambda \rightarrow$

$\bar{K_i} - \bar{K_f} = Q \quad \frac{4\pi \sin\theta}{\lambda}$

$|\bar{K_i}| \neq |\bar{K_f}|$

$\dfrac{\hbar^2 K_i^2}{2m} - \dfrac{\hbar^2 K_f^2}{2m} = \Delta E$

$Q^2 = K_i^2 + K_f^2 - 2 K_i K_f \cos\theta$

$\omega \uparrow$

$-\pi/a$

O

π/a

$2\pi/a$

$\leftarrow q \rightarrow$

G

$q =$ Phonon momentum

$q =$

$Q =$ Momentum transfer

$G =$ Reciprocal lattice vector

I will go into a little bit more details of the experiments on a triple axis spectrometer. In the beam path (from the reactor), as I showed you at the center of the drum, there is a monochromator crystal. The role of monochromator is to get a monochromatic beam of neutrons from a polychromatic beam. Now, if you consider the reactors flux as a Maxwellian (in wavelength), then the role of this monochromator is that it is a crystal with a known d spacings and we know the Bragg's relation $2d \sin\theta = n\lambda$, so, we choose one specific monochromatic beam by choosing the plane of the monochromator (for Bragg reflection). The angle with respect to the incident beam and the total deviation of the primary beam is 2θ. This is a deviation of the beam with the direct beam that fixes the λ of the monochromatic beam.

Now, I must state a few desirable qualities for a monochromator as I mentioned earlier. I cannot use a perfect single crystal because a perfect single crystal will give a very narrow beam (Darwin width) of reflected neutrons. The low intensity will be a problem. So, what we use actually are known as mosaic crystals. What does a mosaic crystal do? Usually in textbooks, you will find perfect single crystal with parallel planes and you use the Bragg's Law, $2d \sin\theta = n\lambda$ when we assume that this is a perfect single crystal. But in reality, it is different. If this single crystal is strained in a way that this crystal consists of small oriented crystallites like this. So, the crystal is broken into crystallites, which have small orientation angles with respect to each other and then based on relation, $2d \sin\theta = n\lambda$, if this angle θ varies a little then we will also have a spread in λ which is desirable. So, instead of choosing a very narrow beam from the incident beam neutron beam, we try to choose something which is broader in wavelength λ and also on angle.

But now, the question comes that by doing this I have improved the intensity but compromised the resolution as now, our wavelength has got a spread, $\lambda \pm \Delta\lambda$. In this, I can also mention another phenomenon known as second order contamination. We write $2d \sin\theta = n\lambda$ and for example, suppose I am taking (1, 1, 1) reflection from a copper monochromator, now for almost the same angle the (2, 2, 2) reflections add $\lambda/2$ wavelength which will contaminate the reflected beam. There are various ways that I can get rid of this contamination such as we can put a filter in the beam which will cut down the $\lambda/2$ i.e. more energetic neutrons and allow the primary neutrons to go in. Briefly, we want a broader beam with wavelength spread but get rid of the contamination due to second order reflection using a filter. The mosaic spread of the monochromator will give me additional neutron intensity and of course, I make a little bit of compromise on resolution.

Now, the analyzer which analyses the energy of the outgoing beam by choosing reflection from a Bragg plane, is sometimes pyrolytic graphite. This plane is chosen at an angle, you can say θ analyzer, then this wavelength λ Bragg-reflected by the analyzer is known, because of the angle.

Everywhere there are contributions from instrumental resolution which is mosaic spread. Ultimately these two mosaic from monochromator and analyzer will sum up in square. The fact is that broader wavelength will give me better intensity to do the experiments. So, now, I have is 2 changes in the scattering, one is $K_i - K_f = Q$ and this $Q \neq \frac{4\pi \sin \theta}{\lambda}$. I must mention it specifically, because now it is an inelastic process. So, length of K_i is not equal to length of K_f, $|K_i| \neq |K_f|$ and $\frac{\hbar^2 K_i^2}{2m} - \frac{\hbar^2 K_f^2}{2m}$ is the energy difference between the two beams, incident and the scattered. Above is the energy transfer in the inelastic experiment and

$$Q^2 = K_i^2 + K_f^2 - 2|K_i||K_f|\cos\theta$$

if θ is the angle between K_i and K_f. These are the equations which will be valid for an inelastic neutron scattering experiment and then we have to arrange our sample, analyzer and the detector in a way that either we can perform the scan along a path is Q-E or Q-ω space, where Q is the momentum transfer, E or $\hbar\omega$ is the energy transfer. It can follow a path in a constant q (phonon wavevector) or in a constant E (ω) with respect to phonon dispersion relation. If you remember, earlier I have shown you the phonon dispersion relation somewhat like this. This is ω versus q. So, I can do this scan along the constant q path or I can do the scan along the constant energy path (ω).

Again, I must caution you that I have got three definitions for momentums. One is q, the phonon momentum and when I am plotting ω versus q, this is phonon wavevector and this is limited between $-\frac{\pi}{a}$ to $\frac{\pi}{a}$ for a linear lattice of length a. So, if I go to $\frac{2\pi}{a}$, that gives me one reciprocal lattice vector G but I can always reflect back anything I have got beyond $\frac{\pi}{a}$ from here to here (inside Brillouin zone). Just by subtracting $\frac{2\pi}{a}$ from this value, I can bring it back to the first Brillouin zone. I will show you the experimental results later, that we do the plot in first Brillouin zone for some symmetry direction in momentum space, in second Brillouin zone for another symmetry direction, in third Brillouin zone for another symmetry direction. The phonon dispersion relations in a crystalline lattice depends on the direction of propagation of

the phonon or the direction of the wave vector q. There are specific symmetry directions in a crystal along which the phonon dispersion relations will change depending on the direction and all of them needs to be found out experimentally.

So, I have got a q which is phonon wavevector or as I said phonon momentum vector, Q which is momentum transfer in the experiment and G the reciprocal lattice vector. So, before we start or even plan an experiment for phonon dispersion relation, we must know the crystallographic structure and we need a single crystal for experiments to obtain phonon dispersion relations, because phonon dispersion relations can be different along different symmetric directions in the crystal and for that we need a single crystal which we can orient for experimental purpose.
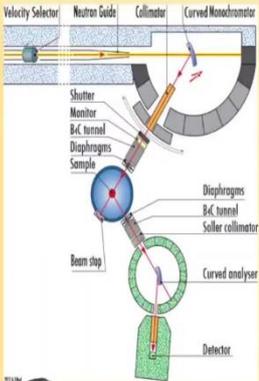
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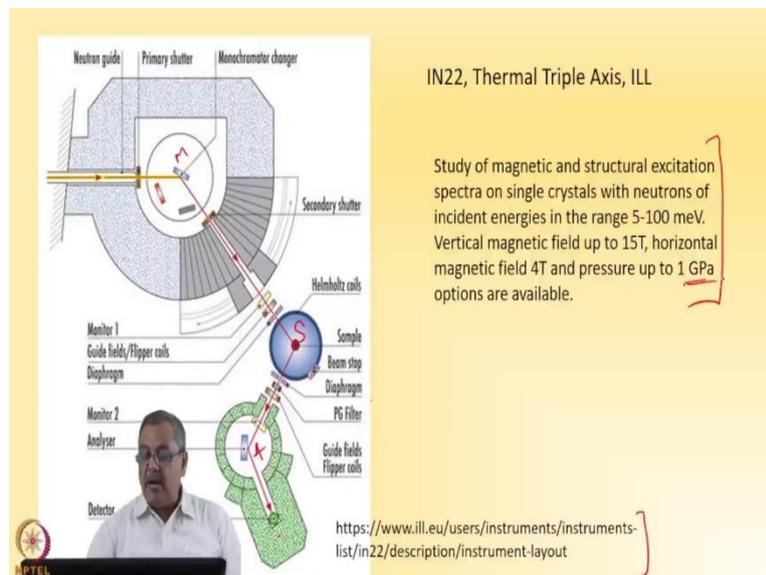
Let me show you other spectrometers at major neutron sources. I tried to show the triple axis spectrometers from various nuclear reactors. Looking at spectrometer at IN22 beamline at ILL, the design is similar, this is the monochromator, here is the sample and there is an analyzer on the third axis and then there is detector. So, the basic structure is same, but what are added advantages of a powerful neutron source? I have taken the parameters from the online description of the instrument at ILL, Grenoble, France.

You can see that at IN22, ILL the incident beam can have incident energies in the range of 5 to 100 meV. A reasonably large energy range can be covered. It has got a vertical magnetic field up to 15 T, horizontal magnetic field up to 4 T and we can also do high pressure experiments up to 1 GPa. Because phonons are the collective oscillation of the atomic lattice

sites, with pressure there are lots of changes because the interatomic distances change and the phonons have difference in their dispersion.

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IN22, Thermal Triple Axis, ILL

Study of magnetic and structural excitation spectra on single crystals with neutrons of incident energies in the range 5-100 meV. Vertical magnetic field up to 15T, horizontal magnetic field 4T and pressure up to 1 GPa options are available.

$$Q = (Q_x, Q_y, Q_z)$$
$$(Q_x, Q_y)$$
$$Q$$

https://www.ill.eu/users/instruments/instruments-list/in22/description/instrument-layout

Similarly, there is another three-axis spectrometer for cold neutrons, IN12 at ILL. Because it is a cold neutron spectrometer, we can do lattice dynamics at low frequencies or measure low energy phonons. When I have cold neutrons that means, typically the energies are below 5 meV and wavelength is longer than 4 Å. Such an instrument needs a cold neutron source installed in the reactor. That is why, this is an added advantage here at ILL Grenoble because they have a cold neutron source. You can use the cold neutrons in ILL, for studying slower dynamics. Lattice dynamics at low frequencies indicate dynamics, critical scattering etc. ILL also can probe a weak static magnetic field. Here, we can study magnetic multilayers and thin films.

Because of slower dynamics, most of the biological model membranes can be studied here as their dynamics are extremely slow. Also, magnetic excitations like magnons can be measured using this spectrometer. But the basic structure of the three instruments that are described here, the triple axis spectrometer at Dhruva, the IN22 thermal triple axis at ILL and other spectrometer IN12 at ILL, are similar. Here you can see, because I am working with at low energies, the monochromator is curved means you have the curvature like this. This curved monochromator will tend to focus the beam; hence, the beam dimensions will be small but neutron intensity will increase at the cost of increased divergence (or resolution).

So, at the cost of divergence or resolution, I can get better intensity. You can also have vertically focused monochromator. But experiment is usually done in the horizontal plane. In that case, you can gain intensity, but you will not lose resolution in the horizontal plane. So, if you see this photograph, these are very heavy equipment like detector or rotation stages, all moving in horizontal plane. So, almost always, since I do not know of any instrument doing an experiment with a vertical geometry, the experiment is done in horizontal geometry. All the

instruments that I showed, it is always horizontal geometry. That means Q which in general should be having three components (Q_x, Q_y, Q_z), if I consider z is the vertical axis, then generally Q is (Q_x, Q_y). Playing with the angle between the monochromator and the angle at which we put the analyzer, we can decide what Q values I am choosing, and I can do a constant Q scan.

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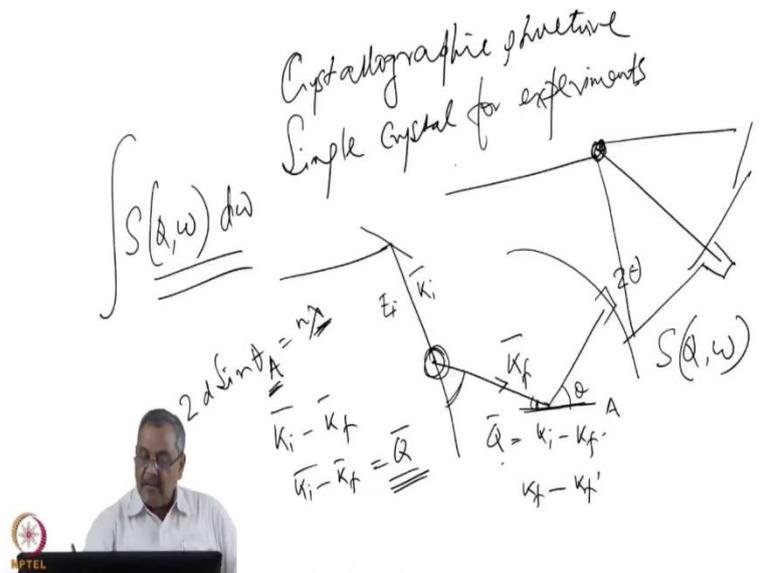
Assume this is the incident energy and this is the wavevector incident on the sample. Now, this angle decides what is the wave vector transfer Q. I have got an analyzer here and the detector rotates around it in θ-2θ mode. So, at this analyzer angle when I rotate the analyzer and this is θ, and I rotate the detector 2θ. So, these are coupled and once we couple them for a Bragg peak, they remain coupled, but this K_f changes. So, K_f goes to K_f' because this angle has changed. Now, from this beam you are taking another neutron whose energy is slightly different because this angle is different. Because at analyzer, using $2d \sin \theta_A = n\lambda$, when I increase the θ_A, I increase the λ. That means K_f' is shorter because λ is longer now.

Now, I have another $K_i - K_f$ at this angle and I can adjust these angles to keep $K_i - K_f$ at a fixed Q which is a momentum vector transfer in this experiment. Earlier when we did structural work, it was much simpler, I had a following it either I had a detector in end-on geometry here or I had a position sensitive detector which covered the whole Q range. And at that time, we were probing the structure, which I explained to you earlier with integrating the scattering law over energies $S(Q) = \int S(Q, \omega) d\omega$ and we were not bothered about the outgoing energy. Now, we are trying to find out $S(Q, \omega)$ and that is why all this trouble with the analyzer and its setting.