Neutron Scattering for Condensed Matter Studies

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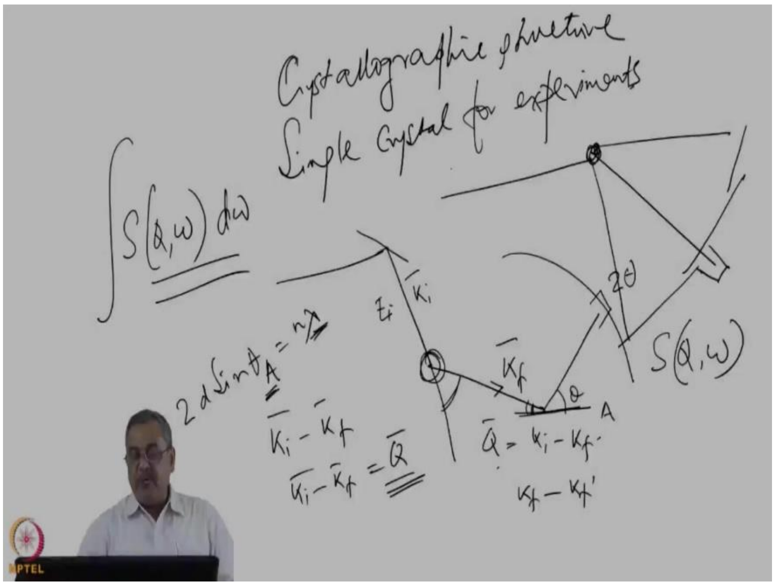

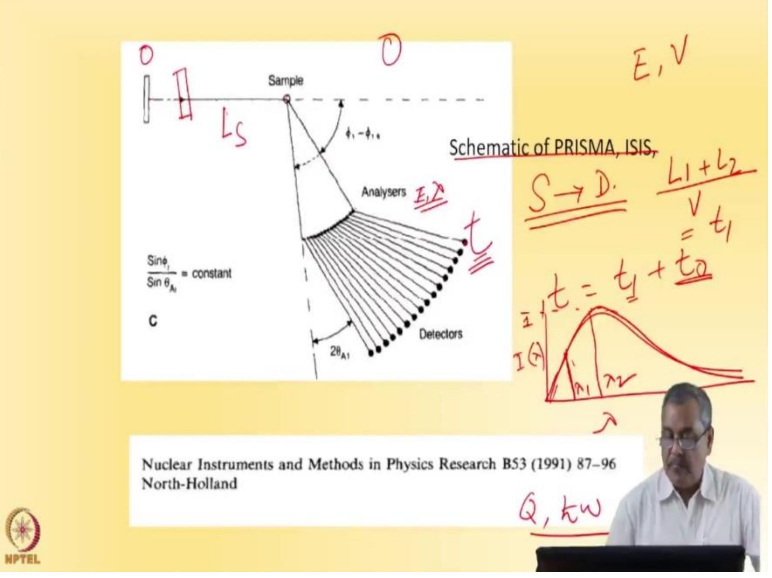

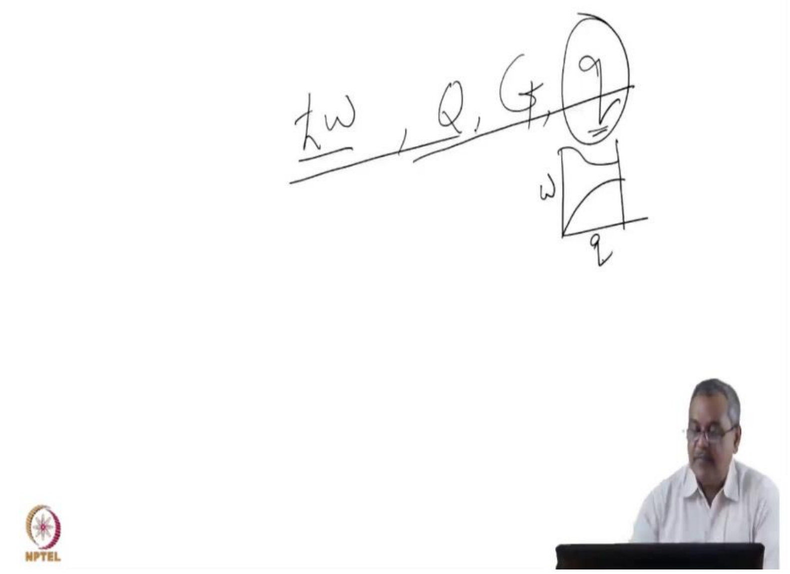

Next, I will talk to you about an experimental setup in a spallation neutron source which uses a polychromatic incident beam

This is a schematic of PRISMA instrument at ISIS. Here, we use time of flight (ToF) technique. Please note that we have a sample at a certain distance, say L_s from the source and then I have a bank of analyzers here at a certain distance from the sample with varying angles of reflection and then the detector bank.

So, what the instrument does actually? You can see that starting from here (the source) if this is the 0 time, at a time '*t*' a neutron is detected. Now, knowing this time interval '*t*', we know, what is the energy or wavelength of the neutron that I am detecting in this detector, as I know at what time the neutron is detected. That means, I can calculate the energy/wavelength because I can determine the wavelength or energy from velocity of the neutron. If sample to analyzer distance is L_1 and analyzer to detector distance is L_2, so, total distance covered by neutron here is $L = L_1 + L_2$ dividing it by time 't' gives the velocity 'v' of the neutron. Once I know the time taken for this path, I can find out the time taken from the source to the sample.

By zero time I mean when I start the clock. Usually, in a spallation neutron source, the neutrons are produced by impinging protons on a target of high Z material which produces neutrons by spallation in a very short span of time with respect to the timescales that I am talking about. That is time for moderation of the neutron (bringing the energy down from few or tens of MeV to meV) and the time for distances covered. When the proton beam hits the spallation target, I start my clock and that is time 0. And when it is detected in this detector that is time t. Now, knowing this t, I can calculate out what is the t_1, let us say for the given energy/wavelength for this distance $L_1 + L_2$ time taken is t_1. Then I can calculate out what is the time taken from the source to the sample t_0 and once I know this, I can find out what is the wavelength of that neutron which has reached the detector. In this case, we do not have a monochromator, so, in case of a spallation neutron source, what is incident on the sample is a polychromatic beam.

So, from the time of flight, I can find out what is the wavelength of the neutron that has reached the detector and just now I described to you that once I know the outgoing energy, so, I can find out the time taken for the total path length. From the total time then I can find out what is the t_0 and from that I can find out what is the wavelength of the neutron.

Now, because the incident neutron flux is a Maxwellian distribution in energy, the intensity of neutrons with different wavelengths is not same. So, there has to be a monitor detector here, which finds out the intensity of the incoming neutrons for normalizing scattered neutron data. In the scattered intensity we have to weigh each and every neutron with respect to this weightage in the incident spectrum . Because neutrons falling on the sample with different wavelengths do not have same intensity, hence, we have to scale them as per their intensity in the scattered beam. That means, if I scale the maximum intensity to one, then for example, this intensity at any other wavelength will be like one divided by some fraction epsilon ($1/\varepsilon$), which will scale up this intensity at that wavelength because that one has smaller number of neutrons compared to the wavelength λ corresponding to maximum intensity. In this way, normalization is done corresponding to all the wavelengths. Time of flight (ToF), I can find out as described above followed by λ of the scattered intensity from ToF. So, I can record the whole scattered intensity in terms of momentum transfer Q and energy transfer $\hbar\omega$.

This is the structure for spallation neutron source for doing time of flight study using inelastic neutron scattering. When we do diffraction experiment here, then this (energy) analyzer will be absent. And then again, this part of scaling will remain same, but we will have the total time and that I will convert it to λ for the incoming neutron.

I discussed this PRISMA spectrometerat the spallation neutron source ISIS at RAL, UK because I will show you a data which has been done in Dhruva reactor, in ILL in France, and then in ISIS at Rutherford Appleton Laboratory, UK. And the entire data was ultimately stitched together to give the phonon dispersion relations. I will come to that later.

So, the fact remains that measuring phonon dispersion relation, has a different order of difficulty compared to if I talk about powder neutron diffraction or even single crystal neutron diffraction. Because in these experiments, we need to be very careful about the incoming energy, the outgoing energy and how to this determine the $\hbar\omega$ for a given momentum transfer (Q), for a given reciprocal lattice vector (G), for a given phonon momentum vector (q). Ultimately, this is the thing (phonon dispersion relation) we want to find. We want to find the dispersion relation as a function of q. This is what your aim is and for that, all these we have to find out.

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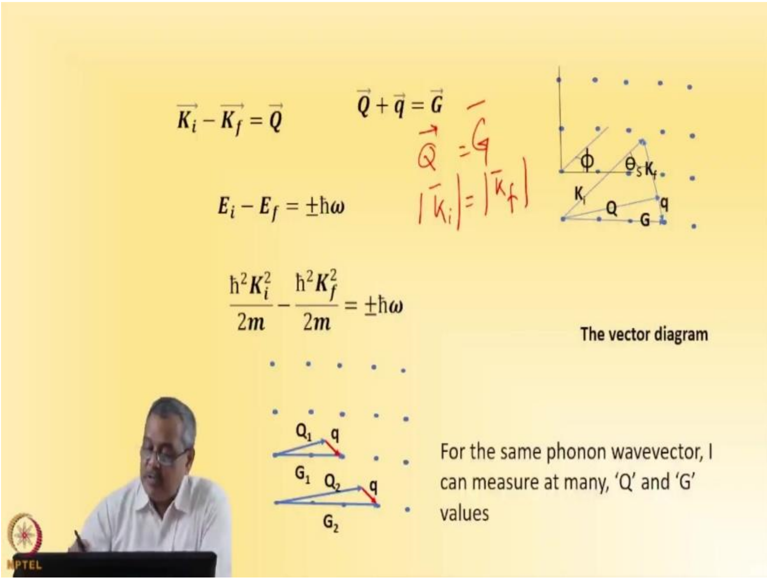

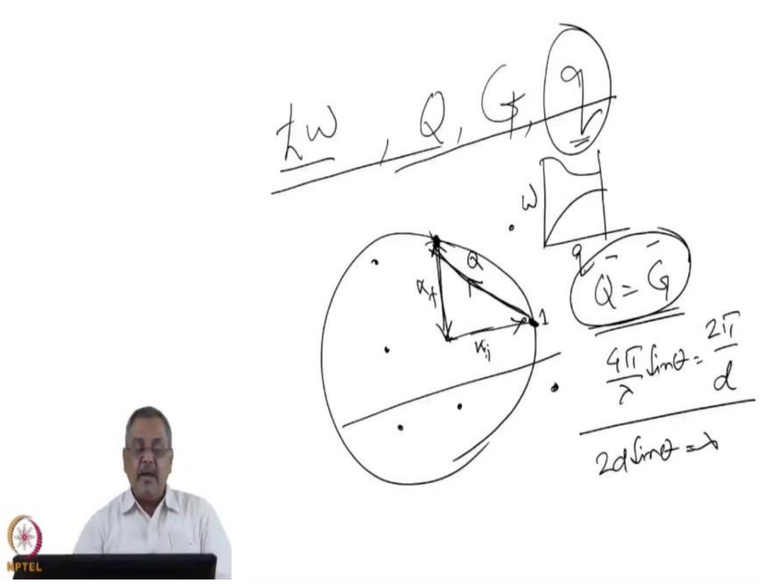

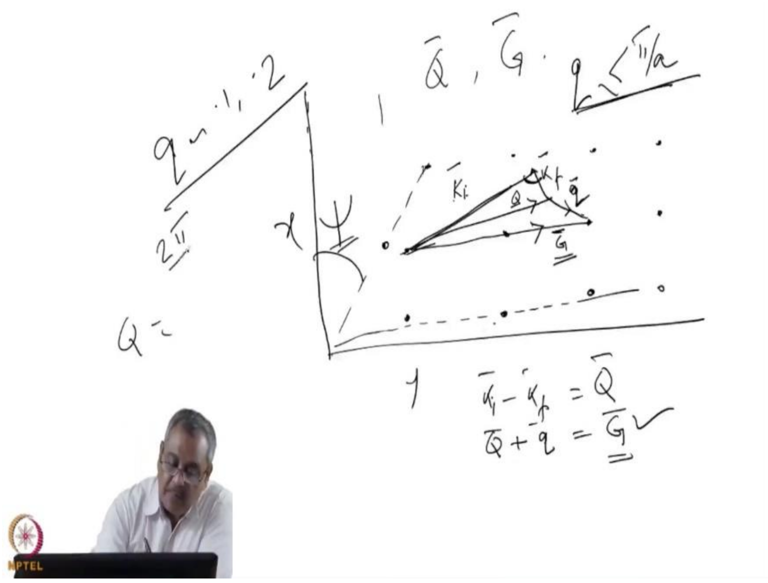

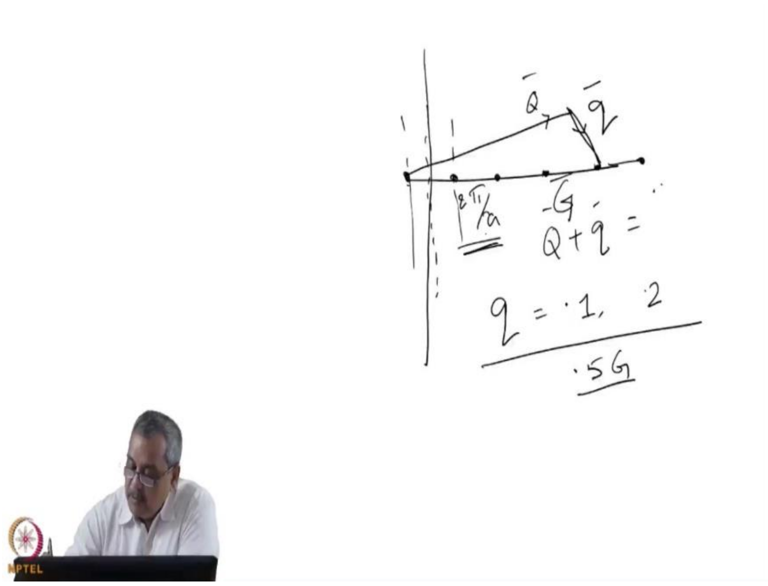

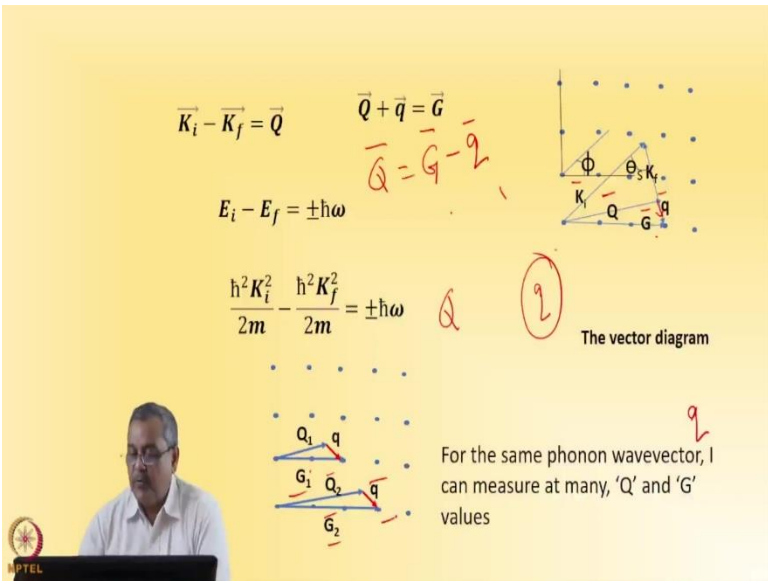

$\vec{K_i} - \vec{K_f} = \vec{Q}$
$\vec{Q} + \vec{q} = \vec{G}$
$E_i - E_f = \pm\hbar\omega$
$\frac{\hbar^2 K_i^2}{2m} - \frac{\hbar^2 K_f^2}{2m} = \pm\hbar\omega$
The vector diagram
For the same phonon wavevector, I can measure at many, 'Q' and 'G' values

When we discussed about diffraction experiment for structure, we wrote momentum transfer should be equal to a reciprocal lattice vector. That is what the Ewald plot was all about. And also, for a diffraction experiment, $|K_i| = |K_f|$, because there is no energy transfer. Ewald construction: if this was a reciprocal lattice vector, if you remember, I start drawing a sphere of radius *K*, which ends on a reciprocal lattice point. I am drawing a circle with this *K* as the radius. And when I do that, if I have this circle or 3-dimensional sphere, intersecting another reciprocal lattice point, then you can see if this is K_i and this is K_f, then you can see $K_i - K_f$ will be equal to the *G*, a reciprocal lattice vector. In this case because this circle has intercepted another reciprocal lattice point that means *Q* = *G*, which is the Bragg law in another form. If I say $Q = \frac{4\pi \sin\theta}{\lambda}$ then from this relation we have $\frac{4\pi \sin\theta}{\lambda} = \frac{2\pi}{d}$ or $2d \sin\theta = \lambda$. So, *Q* = *G* is a more fundamental way of saying that this is satisfying Bragg's Law and I will have a reflected beam in this direction. This was Ewald construction.

Presently, I also have got a phonon associated with the scattering. So, now my rules change a little bit. Earlier, I had *Q* = *G*. Now, Q is equal to the reciprocal lattice vector (*G*) minus a phonon momentum vector (*q*) which is the signature of the phonon dispersion. So, let me now draw it once again for the case of phonons.

Let us say, this is the reciprocal lattice vector and these are my axis let us say *x* and *y*, so that my reciprocal lattice is oriented at some angle (ψ) with respect to the chosen directions. Now, this ψ is basically corresponding the orientation of crystal with respect to the sample setup. Let us say this is $\boldsymbol{K_i}$ the incident vector, this is $\boldsymbol{K_f}$ the final wavevector and then this is the vector $\boldsymbol{Q}$. So, vectorially $K_i - K_f = Q$, $K_i - K_f$ is a momentum transfer *Q*.

Now, this is a reciprocal lattice vector ***G***, and $\boldsymbol{G} - \boldsymbol{Q} = \boldsymbol{q}$ or $\boldsymbol{Q} + \boldsymbol{q} = \boldsymbol{G}$ the reciprocal lattice vector. This is the selection rule for inelastic neutron scattering in a crystal lattice.

Please note there is an angle between $\boldsymbol{K_i}$ and $\boldsymbol{K_f}$, which is the angle between the incident direction and outgoing direction which dictates momentum transfer $\boldsymbol{Q}$ and this has to be associated with a phonon wave vector $\boldsymbol{q}$ in some branch of phonon dispersion curve and $\boldsymbol{Q} + \boldsymbol{q}$ should be equal to **G** the reciprocal vector in this diagram.

Now, if I consider this diagram, this is a reciprocal lattice vector and this is not in the first Brillouin zone. I can go over several Brillouin zones depending on my $\boldsymbol{Q}$ value and what $\boldsymbol{G}$ value I am choosing. Only this *q* (the phonon wavevector) is less than or equal to π/a. So, *q* can be 0.1, 0.2 of the reciprocal lattice vector. Now, the reciprocal lattice vector is $2\pi/a$ long. Let me just repeat what I mean to say, if this is my reciprocal lattice, a linear chain here, then here this $2\pi/a$ is the separation between neighboring reciprocal lattice points and q is less than π/a.

So, when I say q is equal to let us say 0.1, 0.2, it is a fraction of reciprocal lattice vector. This is a multiple of $2\pi/a$ by either 0.1, 0.2 and so on, but it has to be less than 0.5G, because at 0.5G is the boundary of the Brillouin zone comes and q should be less than that for our choice. So, when I am choosing my q, I can go several reciprocal lattice vectors, then this is the phonon vector q.$\boldsymbol{Q}$ plus $\boldsymbol{q}$ should be equal to some reciprocal lattice vector $\boldsymbol{G}$ for the experiment.

Here, I have shown it in the sketch that this was $\boldsymbol{K_i}$ and this angle ϕ (or I ψ in my previous drawing) is the angle of the reciprocal lattice with respect to the chosen coordinate system. That means the sample axis the second axis, can be rotated to give me this ϕ. If this is my incident wavevector $\boldsymbol{K}_i$ and this is vector $\boldsymbol{K_f}$ the final wavevector, angle between them is θ, this is chosen by choosing the angle of scattering and the analyzer position. So, it is chosen according to the sample at what angle I want to put the analyzer to detect the neutrons. After that, of course, neutrons will scatter further and go to the detector.

So, this vector $\boldsymbol{Q}$ and this is a small wave vector $\boldsymbol{q}$, together they should be equal to a reciprocal vector $\boldsymbol{G}$., In this process, for phonon wavevectors which is $\boldsymbol{q}$, I can measure many $\boldsymbol{q}$ at many $\boldsymbol{G}$ values and add them up. I have just shown a general relationship here. I can measure $\boldsymbol{Q}_1$ a momentum transfer for this $\boldsymbol{q}$ value with the reciprocal $\boldsymbol{G}_1$ or I can measure the same $\boldsymbol{q}$ even on a longer momentum transfer $\boldsymbol{Q}_2$ for another reciprocal lattice vector $\boldsymbol{G}_2$. I am telling you repeatedly, the $\boldsymbol{Q}$, I can and change them to get various phonon wavevectors and my dispersion relation is plotted against this $\boldsymbol{q}$.

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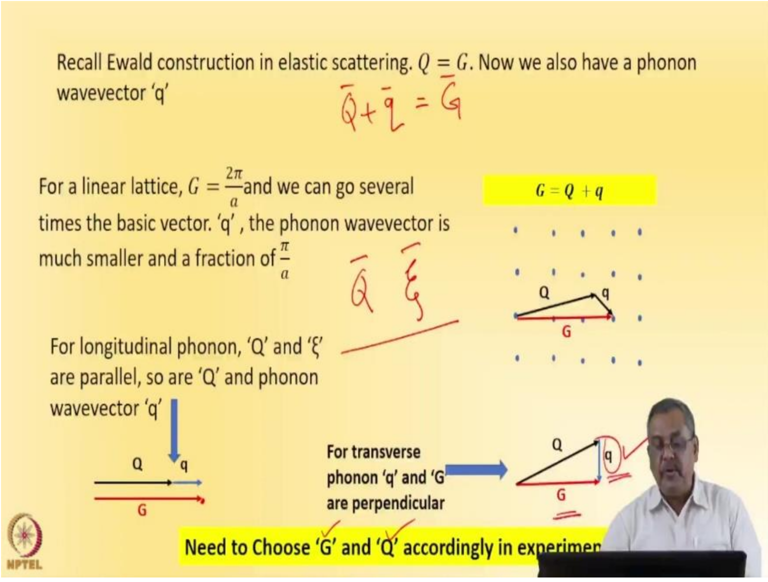

Let again recall the Ewald construction that we had discussed earlier when $\boldsymbol{Q}$ was equal to $\boldsymbol{G}$ for diffraction experiments. If I also have phonon momentum vector $\boldsymbol{q}$ and then for momentum conservation it is $\boldsymbol{Q} + \boldsymbol{q} = \boldsymbol{G}$. These are vector terms. For a linear lattice $G = 2\pi/a$ and we can go several times the basis vector to do a measurement at a certain $\boldsymbol{Q}$ as shown.

Now consider a longitudinal phonon. In longitudinal phonon, $\boldsymbol{Q}$ *and* $\boldsymbol{\xi}$ are parallel, where $\boldsymbol{Q}$ is momentum vector transfer and $\boldsymbol{\xi}$ is the displacement of the atom. The geometry looks like this. That means, here they are on the same line and $\boldsymbol{Q} + \boldsymbol{q} = \boldsymbol{G}$.

Similarly, for a transverse phonon the displacement is normal to the $\boldsymbol{G}$ values, and it should be looking like this where the $\boldsymbol{G}$ and $\boldsymbol{q}$ are normal to each other. Because this is the reciprocal lattice vector and is similar to what we have in real space. The directions remaining same, the phonon wavevector or the displacement of the atoms are normal to the reciprocal wave vector that is how this diagram shows that $\boldsymbol{G}$ and $\boldsymbol{q}$ they are perpendicular.

So, in every experiment, before you do the experiment, we have to choose the $\boldsymbol{G}$ values and the $\boldsymbol{Q}$ values and then that will dictate what will be the $\boldsymbol{q}$ value in our measurements. There is more to it, and there are symmetry relations. I will come to it in the next portion of the topic.