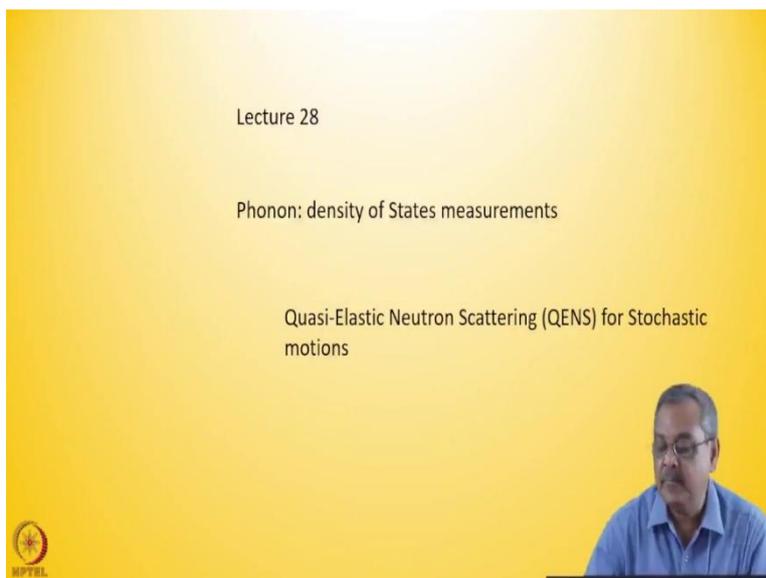


**Neutron Scattering for Condensed Matter Studies**  
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**Homi Bhabha National Institute**  
**Week 11: Lecture 28A**

**Keywords: Phonons, Density of states, Dispersion relation, Debye Waller factor, Stochastic motion**

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I had been discussing phonons, the collective oscillation of atoms and molecules in a crystalline solid. So far, I have gone in the discussion about how to measure the phonon dispersion relations in material and that deals with single crystal samples. Today, I will go ahead and talk about phonon density of state measurements, which is done with powder samples, as powder samples are more easily available and for many physical phenomena density of states is good enough to give us an answer. Then, I will stop dealing with phonons and I will go into quasi-elastic neutron scattering for stochastic motions.

So, I will discuss two different kinds of motions. Phonons where we have collective periodic oscillations and their time correlations are known. So, this is a coherent effect. We found out crystallography structure using coherent neutron scattering cross section. Phonon dispersion relations are also found out using coherent scattering cross section. Whereas, quasi-elastic neutron scattering is for stochastic motions. Stochastic means statistical motions (not coherent). They are more in the realm of diffusion; when a molecule diffuses statistically in a liquid or even in some geometrical confinements.

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**Single crystal:**  
Gives information about  $\omega(q)$   
Good quality single crystals (1 cc) are not easily available.

**Polycrystalline sample:**  
Provides phonon density of states  $g(\omega)$   
Sample of about 10 cc.

For single crystals, we get information about  $\omega$  vs  $q$  and determining it is an involved process as I discussed with you so far. These are the dispersion relations for longitudinal and transverse acoustic modes, and optic modes. They have to be determined at  $q$  values inside the 1<sup>st</sup> Brillouin zone. We can do the measurements with a range of choice of  $G$  (reciprocal lattice vector) and  $Q$  (momentum transfer) for a single crystal. I have discussed these issues and towards the end of the previous talk, I had shown you results of measurements on a Zircon Crystal. Zircon is the constituent of the earth's minerals and I showed you that it needs a involved calculation of the phonon dispersion relations in which we also need to use group theoretical considerations where various group theoretical classes or representations are measured at different  $G$  and  $Q$  values. Now, from there we go into polycrystalline sample. Polycrystalline samples are easier to get since these are powder samples. They provide phonon density of states and we can work with a sample typically about 10 cc volume.

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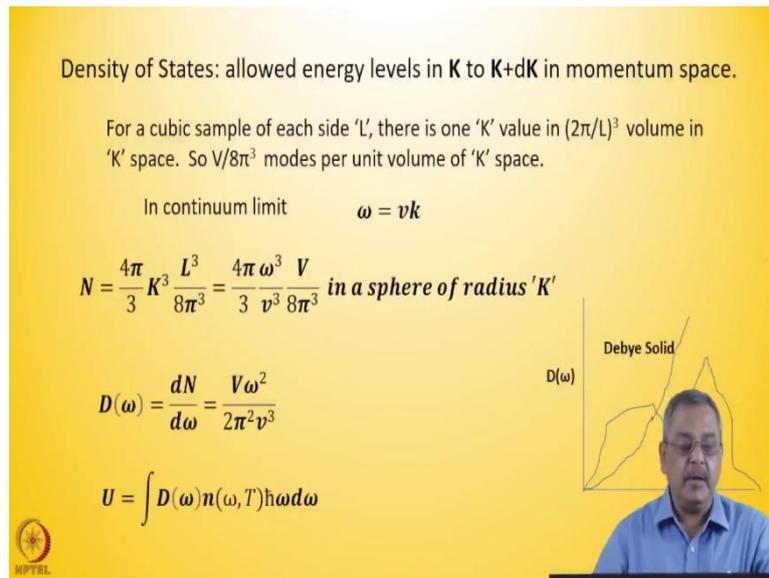
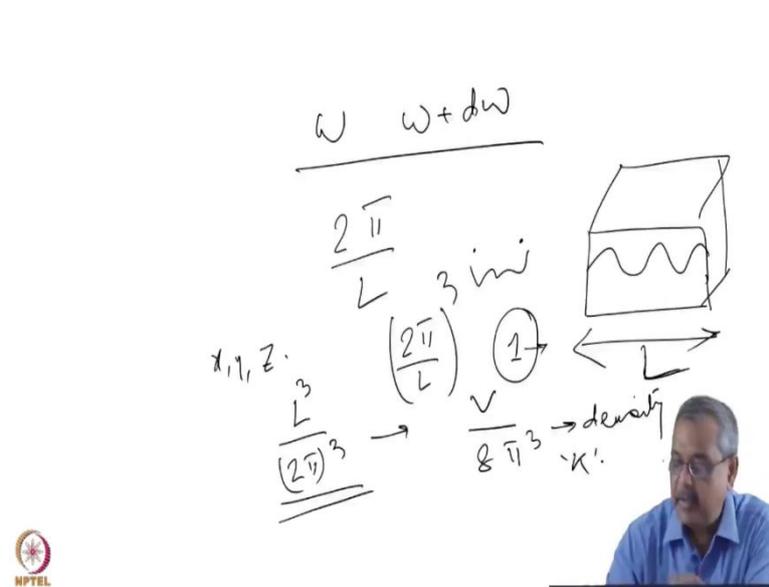
Density of States: allowed energy levels in  $\mathbf{K}$  to  $\mathbf{K}+d\mathbf{K}$  in momentum space.

For a cubic sample of each side 'L', there is one 'K' value in  $(2\pi/L)^3$  volume in 'K' space. So  $V/8\pi^3$  modes per unit volume of 'K' space.

In continuum limit  $\omega = vk$

$$N = \frac{4\pi}{3} K^3 \frac{L^3}{8\pi^3} = \frac{4\pi \omega^3}{3 v^3} \frac{V}{8\pi^3} \text{ in a sphere of radius 'K'}$$

$$D(\omega) = \frac{dN}{d\omega} = \frac{V\omega^2}{2\pi^2 v^3}$$

$$U = \int D(\omega) n(\omega, T) \hbar \omega d\omega$$



Handwritten notes on a whiteboard explaining the density of states. It shows the derivation of the density in  $K$ -space as  $\frac{V}{(2\pi)^3}$ . A diagram shows a 3D box with a wavy line representing a wave inside, with a double-headed arrow indicating length  $L$ . The notes include the expression  $\left(\frac{2\pi}{L}\right)^3$  and a circled '1'.

Now, what do we mean by density of states? That is the number of states we have between  $\omega$  and  $\omega + d\omega$ . I will quickly remind you about it what we did in our master's degree. Suppose, I have a crystal which is having (assumed) dimensions  $L$  (a cube) where  $L$  is quite large, a very large dimension. So, for that I know quantum mechanics demands that in one dimension I have a gap  $\frac{2\pi}{L}$  in momentum space between two allowed 'K' values, because there should be a stationary wave inside the sample. And if I consider three-dimensional momentum space then the volume  $\left(\frac{2\pi}{L}\right)^3$  will have one allowed  $K$  value then the density of states will be  $\left(\frac{L}{2\pi}\right)^3$ . This goes to  $\frac{V}{(2\pi)^3}$ . This is the density in 'K' space.  $V$  is the sample volume here.

Please remember, here I drew in a real space, but this is the density in  $k$  space. So, in  $K_x, K_y, K_z$  direction if I talk about a sample of size  $L$ , then in a unit volume in ' $K$ ' space density will be  $\left(\frac{L}{2\pi}\right)^3$  because  $\left(\frac{2\pi}{L}\right)^3$  is the volume in which we have got one allowed  $K$  value.

(Refer Slide Time: 06:05)

The image shows a handwritten derivation of the density of states  $N(\omega)$  for a linear dispersion relation  $\omega = vk$ . The derivation starts with the volume of a sphere in  $k$ -space,  $\frac{4\pi}{3}k^3$ , and then converts it to  $\omega$ -space using the relationship  $k = \omega/v$ . The final expression for the density of states is  $N(\omega) = \frac{4\pi}{3} \frac{\omega^3}{v^3} \frac{V}{8\pi^3}$ , and the differential form is  $dN(\omega) = \frac{4\pi}{3} \frac{3\omega^2}{v^3} \frac{V}{8\pi^3} d\omega$ . The derivation also includes a diagram of a sphere in  $k$ -space and a graph of  $\omega$  vs  $k$  showing a linear relationship.

I know that in the lower limit when  $\omega$  vs  $K$  is linear (for a sound wave) then  $\omega = vK$  where  $v$  is the velocity of the wave. In that case, in the  $K$  space, if I consider a sphere of radius  $K$ , then it has got a volume of  $\frac{4\pi}{3}K^3$  and in this volume I need to find out how many allowed  $K$  values will be there. It will be,  $\frac{4\pi}{3}K^3 \times \frac{V}{(2\pi)^3}$ . Now, I can substitute in this the relationship between  $\omega$  and  $K$  i.e.  $K = \omega/v$  where  $v$  is the velocity of the wave. Here it is sound velocity when I talk about  $K \rightarrow 0$  limit in case of phonons. I am giving a simple derivation. It becomes,

$$N(\omega) = \frac{4\pi}{3} \frac{\omega^3}{v^3} \frac{V}{8\pi^3}$$

So that,

$$dN(\omega) = \frac{4\pi}{3} \frac{3\omega^2}{v^3} \frac{V}{8\pi^3} d\omega$$

$$dN(\omega) \propto \omega^2 d\omega$$

This is a for a dispersionless linear relationship for  $\omega$  vs  $k$  where density of states goes as  $\omega^2$ . And we know that this is true for free particles also.

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Density of States: allowed energy levels in  $\mathbf{K}$  to  $\mathbf{K}+d\mathbf{K}$  in momentum space.

For a cubic sample of each side 'L', there is one 'K' value in  $(2\pi/L)^3$  volume in 'K' space. So  $V/8\pi^3$  modes per unit volume of 'K' space.

In continuum limit  $\omega = vk$

$$N = \frac{4\pi}{3} K^3 \frac{L^3}{8\pi^3} = \frac{4\pi \omega^3}{3 v^3 8\pi^3} V \text{ in a sphere of radius 'K'}$$

$$D(\omega) \frac{dN}{d\omega} = \frac{V\omega^2}{2\pi^2 v^3}$$

$$U = \int D(\omega) n(\omega, T) \hbar \omega d\omega$$

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$$D(\omega) = \frac{dN}{d\omega} = \frac{V\omega^2}{2\pi^2 v^3}$$

$$U = \int D(\omega) n(\omega, T) \hbar \omega d\omega \rightarrow U = \int D(\omega) n(\omega, T) d\omega$$

$C = \frac{2U}{2T}$

In a Debye solid the density of states goes as  $\omega^2$ , but in actual crystalline solids this relationship is not so simple. Here, I wrote,

$$D(\omega) = \frac{dN}{d\omega} = \frac{V\omega^2}{2\pi^2 v^3}$$

and it gives me  $D(\omega) \sim \omega^2$  relationship for density of states  $D(\omega)$  and the internal energy is given as an integration over the density of states number of particles (phonons here) present at that density of states at a frequency  $\omega$  and temperature  $T$  and  $\int N(\omega, T) D(\omega) \hbar \omega d\omega$ , that gives me internal energy,  $U$ , of the system.

But, in case of real solid, we have density of states which might look different. And actually, we need to measure the density of states, which I will show you shortly in case of phonons, to find out the internal energy. For example, if I know the internal energy, then from  $\frac{du}{dT}$ , I can find out the specific heat which is important for many of the phase transitions.

(Refer Slide Time: 10:16)

Incoherent approximation to coherent phonon scattering in polycrystalline sample

For large momentum transfer ' $Q$ ' [ $\gg 2\pi/r$ ] one may consider the relative motions as incoherent or there is no phase relation between them

$$|\vec{Q} \cdot \vec{\xi}|^2 \sim Q^2 \xi^2 / 3$$

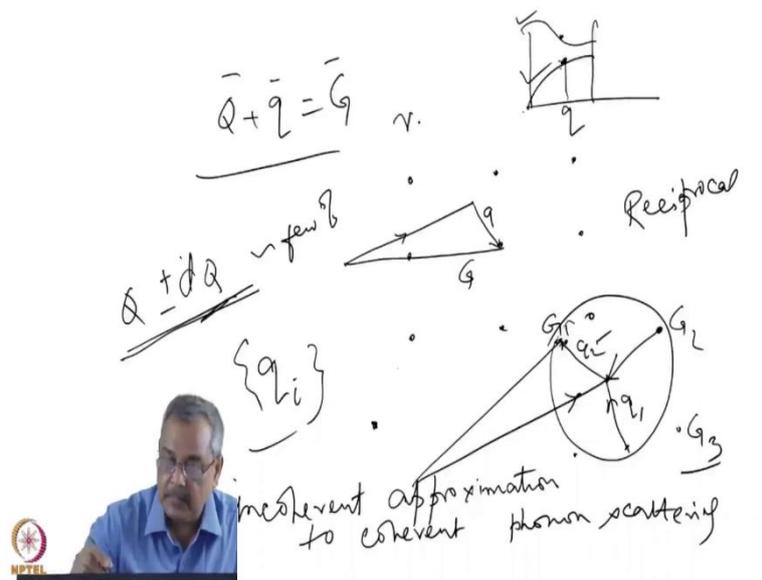
Scattering from 'kth' atom has  $b_{\text{coh}}$  but the motion is incoherent, so we also will add  $b_{\text{incoherent}}$

For many macroscopic thermal information we don't need phonon dispersion relation, but an average information like density-of-states will be sufficient. Example, specific heat.

$$C = \frac{\partial U}{\partial T}$$

Now, what I know is that I have to measure at various  $Q$  (momentum transfer) values. For large momentum transfer when  $Q$  is much greater than  $\frac{2\pi}{r}$ , where  $r$  is the inter particle distance, I can make an approximation that the relative motions are incoherent. This will be clear, if I make a diagram.

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As you remember that earlier also, I had showed you that if this is a reciprocal lattice, then my  $Q$  and  $q$  together should be equal to  $G$  because of the conservation relation,  $Q + q = G$ . Now, there is  $dQ$  uncertainty around  $Q$  that is  $Q \pm dQ$  where  $dQ$  is around few percent. If  $Q$  is large enough, then the resolution ellipsoid, as I showed you earlier, allows contribution from various  $q$  values.

I am just trying to draw it for you; this is a reciprocal lattice. There are various possible combinations of  $G$  values and  $q$  values so that I get a host of  $q$  values i.e. a set  $\{q_i\}$  that are contributing to my  $Q$ . So, I am getting a sum over all  $q$ 's and in the process. Each  $q$  talks about a displacement of atoms in case of phonons.

This is a longitudinal acoustic phonon and this is an optic phonon. Whenever I talk about a phonon, every  $q$  value contributes when I touched a dispersion curve. A certain displacement for the all atoms or one atom. A pattern of displacement for the atoms which are moving coherently, but so many  $q$ 's are contributing to my measurements, then it is a sort of summing over all the  $q$  values.

And now, I can consider that each atom is moving incoherently. That means, I can just simply take the motion of a particular atom without bothering about the correlation with other atoms. This is known as incoherent approximation to coherent phonon scattering.

I am imposing this approximation, and correctly so, because I have got a  $Q$  which is large enough so that the  $\Delta Q$  encircles so many  $q$  values and all the  $q$  values are added together. Each

$q$  value corresponds to a displacement for one particular atom and will give a part of the displacement which I consider. The total displacement of that particular atom coming from all the phonon branches and they are adding up incoherently. Actually, earlier also we had done it, if you remember, in case of Debye Waller factor, we talked about a displacement  $u$  around a mean position  $r$ .

(Refer Slide Time: 14:33)

Handwritten mathematical derivations on a whiteboard:

- $\int_0^\pi \cos^2 \theta d\theta$
- $\underline{\underline{u + r(t)}}$
- $G \cdot r = G r \cos \theta$
- $\langle \cos^2 \theta \rangle = \frac{1}{3}$
- $\underline{\underline{\frac{1}{3} Q^2 u^2}}$

I talked about  $u + r(t)$  in case of Debye Waller factor. And ultimately, when I did that approximation for  $u$  and  $r(t)$  then,  $G \cdot r = G r \cos \theta$ . We found out that square and we showed that if it is spherically isotropic then  $\langle \cos^2 \theta \rangle = 1/3$ . That is very easy to show from the solid angle average from the integration,  $\int_0^\pi \cos^2 \theta d\theta$ . So, it became  $\frac{1}{3} Q^2 u^2$ , if I am talking about any value in  $Q$  space. This was what we got as Debye Waller factor (earlier). In that case also we assumed that each atom is moving independently.

(Refer Slide Time: 15:56)

Incoherent approximation to coherent phonon scattering in polycrystalline sample

For large momentum transfer ' $Q$ ' [ $\gg 2\pi/r$ ] one may consider the relative motions as incoherent or there is no phase relation between them

$|\vec{Q} \cdot \xi|^2 \sim Q^2 \xi^2 / 3$

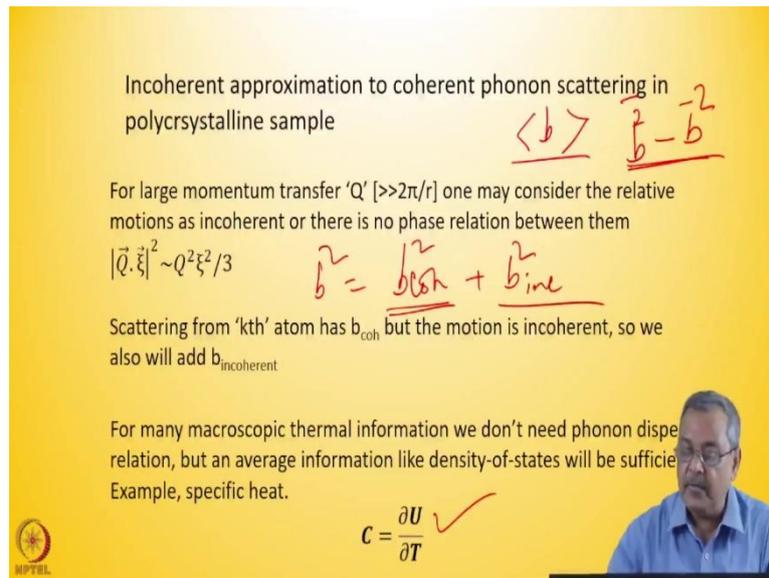
Scattering from ' $k$ th' atom has  $b_{\text{coh}}$  but the motion is incoherent, so we also will add  $b_{\text{incoherent}}$

For many macroscopic thermal information we don't need phonon dispersion relation, but an average information like density-of-states will be sufficient. Example, specific heat.

$c = \frac{\partial U}{\partial T}$

$\langle b \rangle = \bar{b} - \bar{b}^2$

$\bar{b}^2 = \bar{b}_{\text{coh}}^2 + \bar{b}_{\text{inc}}^2$



Here, starting with phonons and then considering the  $Q$  length, we find that for sufficiently large  $Q$  values where sufficiently large means when the resolution in  $Q$  involves adding up of many phonon branches, then that allows me to consider the movement of each and every atom in the lattice as independent though there are actually hidden coherence between the atoms in the lattice when you calculate the phonons. These phonons at large  $Q$ , for each atom, add up and gives me a displacement which is quasi-independent and I can treat them as independent.

I am talking about independent (and incoherent) atom motions. Earlier, if you remember in phonons, my  $b$  (scattering length) was  $b$ -coherent ( $b_{\text{coh}}$ ) only, but now, since the motions are incoherent, so incoherent scattering cross section also will come in. If you remember  $b_{\text{coh}}$  is basically average of  $b$ , ( $\bar{b}$ ), in a lattice. And  $b_{\text{incoherent}}^2 = \bar{b}^2 - \bar{b}^2$ .

Now, instead of using only  $b_{\text{coh}}$ , because I am assuming that the motions are incoherent and independent of each other, my  $b^2$  will comprise  $b_{\text{coh}}^2 + b_{\text{incoherent}}^2$ . So, I get a (scattering cross-section) boost for this. I can put this in my expression and I can get the density of states for phonons. I expressed sometimes back that for many macroscopic thermal information (like specific heat), we do not need the details of phonon dispersion relation, but an average information like density of states will be sufficient.

(Refer Slide Time: 18:30)



$$S_{coh}^1(Q, \omega) = \sum \underbrace{\{f_j^1(Q)\}^2}_{\text{Structure factor}} \left[ \underbrace{n(\omega) + \frac{1}{2} \pm \frac{1}{2}}_{\text{Population factor}} \right] \frac{\hbar}{2\omega} \underbrace{\delta(Q + q - G)}_{\text{Momentum}} \underbrace{\delta(\omega \pm \omega_{q_j})}_{\text{Energy}}$$

$$f_j^1(Q) = \sum_k b_k^{coh} \frac{Q \cdot \xi(q_j, k)}{\sqrt{m_k}} e^{-W_k(Q)} e^{iG \cdot r_k}$$

$$S_{inc}^1(Q, \omega) = \sum_k \frac{b_k^2}{\langle b^2 \rangle} e^{-2W_k(Q)} \frac{Q_I^2}{2m_k} \frac{g_k(\omega)}{\hbar\omega} \left[ n(\omega) + \frac{1}{2} \pm \frac{1}{2} \right]$$

$$g_k(\omega) = \int \sum_j |\xi(q_j, k)|^2 \delta[\omega - \omega_j(q)] dq$$

$g^n(\omega) = A \frac{e^{-2W_k(Q)} \hbar\omega}{Q^2 [n(\omega, T) + \frac{1}{2} \pm \frac{1}{2}] S(Q, \omega)}$

$g^n(\omega) = B \sum_k \frac{4\pi b_k^2}{m_k} g_k(\omega)$



$\underline{\underline{Q}}$       2  
 $\underline{\underline{g(\omega)}}$   
 0




$$S_{coh}^1(Q, \omega) = \sum \underbrace{\{f_j^1(Q)\}^2}_{\text{Structure factor}} \left[ \underbrace{n(\omega) + \frac{1}{2} \pm \frac{1}{2}}_{\text{Population factor}} \right] \frac{\hbar}{2\omega} \underbrace{\delta(Q + q - G)}_{\text{Momentum}} \underbrace{\delta(\omega \pm \omega_{q_j})}_{\text{Energy}}$$

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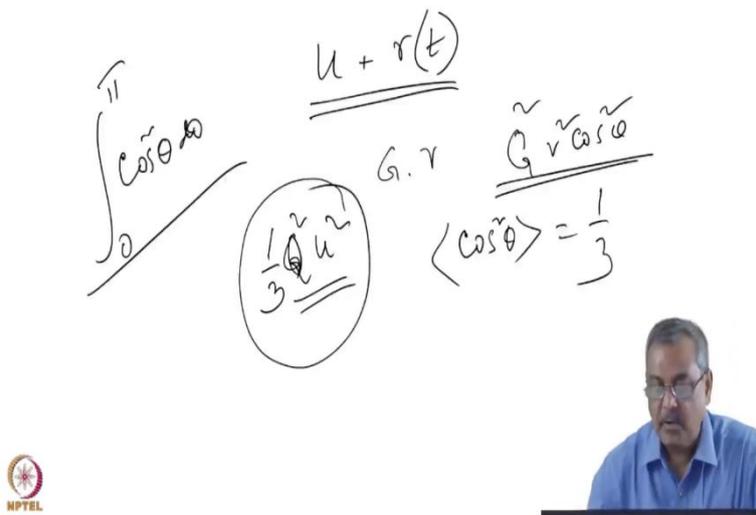
$$S_{inc}^1(Q, \omega) = \sum_k \frac{b_k^2}{\langle b^2 \rangle} e^{-2W_k(Q)} \frac{Q_I^2}{2m_k} \frac{g_k(\omega)}{\hbar\omega} \left[ n(\omega) + \frac{1}{2} \pm \frac{1}{2} \right]$$

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$g^n(\omega) = A \frac{e^{-2W_k(Q)} \hbar\omega}{Q^2 [n(\omega, T) + \frac{1}{2} \pm \frac{1}{2}] S(Q, \omega)}$

$g^n(\omega) = B \sum_k \frac{4\pi b_k^2}{m_k} g_k(\omega)$



Now, how do I estimate the density of states for phonons? I took a simple example earlier, where phonons at  $k$  going to  $0k \rightarrow 0$  as limit  $\omega = vk$ , dispersion less phonon curve that gave as  $g(\omega) \propto \omega^2$ .

Now, let me go back to what I showed you earlier. This expression is a single phonon coherent scattering law. Scattering law will have square of scattering amplitude. So, these are structure factor part. Then, as I explained to you earlier, there is a population factor which gives us the expression for neutron energy gain or energy loss. In case of energy loss, it is  $n(\omega) + \frac{1}{2} + \frac{1}{2} = n(\omega) + 1$  and in case of energy gain,  $n(\omega) + \frac{1}{2} - \frac{1}{2} = n(\omega)$ . Then we had considered a momentum transfer and energy transfer.

Now, here the structure factor has coherent  $b_k^{coh}$  term along with the displacement part where  $Q$  is a momentum vector transfer for the experiment not phonon vector.  $q_j$  is the phonon vector. So, this displacement is for the  $k^{th}$  atom contributed by  $j^{th}$  branch at a momentum vector  $Q$ .

Let me be clearer about it.

I am talking about  $\xi$  and here I am just talking about the acoustic branches. This is the longitudinal branch and this is the transverse branch in the 1<sup>st</sup> Brillouin zone. Now, if I consider this displacement, then I know a displacement pattern for all the atoms will be given. So, if I consider single cell then I know what is the  $k^{th}$  atom and what is its displacement.

Now, for this transverse phonon, I know there will be amplitude in some direction and for the same  $k^{th}$  atom coming from another branch, I have one motion in another direction. That is

what is called  $\xi$  and the dot product of that with  $Q$ , the momentum transfer weighted by the mass of that particular atom. So, if there is an A atom and another atom B in the unit cell and if the  $k^{\text{th}}$  atom is B atom, then the mass of that B atom will be taken here. After that we have terms like a Debye Waller factor and a structure factor.

For the incoherent approximation, I am considering each atom is acting independently, so I need to square the and add the displacements, coming from all the branches of phonons, to get the total displacement. This term will go away and this will be minus  $-2W_k(Q)$ . Now,  $\langle \exp(Q \cdot \chi) \rangle$  I showed you just now, it is  $\exp(-\frac{1}{3}Q^2\xi^2)$  by the same logic by which I used to calculate the Debye Waller factor.  $Q_l^2$  and  $b_k^2$  comes in the expression for the scattering law. Here, I have used it instead of the average  $b^2$  value, this  $b_k^2$  will include incoherent cross-section also. It is  $(b_k^{\text{coh}})^2 + (b_k^{\text{incoh}})^2$ . Under incoherent approximation, because I am considering the motion of the particles are independent of each other, I have used the total scattering cross section incoherent as well as coherent.

Up to this point it was only coherent, but once I have taken the incoherent approximation, my  $f_j(Q)$  will have  $b_k^{\text{total}}$  or  $b_k^2 = (b_k^{\text{coh}})^2 + (b_k^{\text{incoh}})^2$ . I have the expression for density of states here. What is the density of states? If you see the expression here,  $Q^2$  I have been absorbed here. This is  $g_k$  that have  $\xi$  again for  $k^{\text{th}}$  atom, and its contribution to its motion from the  $j^{\text{th}}$  branch at the phonon wave vector  $q$ .

Again, I will repeat, contribution  $k^{\text{th}}$  atom's motion by the  $j^{\text{th}}$  branch at wave vector  $q$  is  $\xi(q_j, k)$  and we square and add contribution from all the branches. , That means if there are three displacements (x,y,z) , say from two transverse phonons and one longitudinal phonon, then the overall displacements will be  $x^2 + y^2 + z^2$ . This I have to do for all the branches, and then I add them up. Keeping the energy conservation means consider the displacements only when we touch one of the phonon branches. And then I have to add up over all the momentum vectors. When I do that, then what I get is density of states for the  $k^{\text{th}}$  atom at phonon energy ' $\omega$ '. And now, if I add up over all the atoms, then what I get is neutron weighted density of states, which is weighted by the scattering cross section of that atom and weighted by the mass of the atom leading to the expression ,  $\frac{4\pi b_k^2}{m_k}$  in density of states for the ' $k^{\text{th}}$ ' atom And then it is  $g_k(\omega)$ , which I get from the displacement of all the atoms.

I have to do an experiment; at large number of  $Q$  values i.e. wave vector transfer. And then I should cover all the branches and I will get  $g(\omega)$  from the experimental measurement. But it is an easier experiment compared to phonon dispersion relation measurements using single crystals. I have given the same thing in terms of an ensemble average of  $S(Q, \omega)$ . Its average is over  $\omega$  here. Similar over  $Q$ , so they are similar, but I like to show you this because what I will get is the sum of  $g_k(\omega)$  in my experiment, rather, I will get  $g^n(\omega)$  in my experiment.

(Refer Slide Time: 25:56)

**Phonon density of states  $g(\omega)$**

$$g(\omega) = c \int_{BZ} \sum_j \delta(\omega - \omega_j(q)) dq$$

Polycrystalline sample

$g(\omega)d\omega$  is the ratio of the number of eigenstates in the frequency interval  $(\omega, \omega + d\omega)$  to the total number of eigenstates.

**Typical phonon dispersion**

**Phonon density of states**

**Partial density of states due to  $k$ -th atom**  
weighted by the component of the eigen-vector

$$g_k(\omega) = c \int_j |\xi(q_j, k)|^2 \delta(\omega - \omega_j(q)) dq$$

$$S_{coh}^1(Q, \omega) = \sum_k \{f_j^1(Q)\}^2 \left[ n(\omega) + \frac{1}{2} \pm \frac{1}{2} \right] \frac{\hbar}{2\omega} \delta(Q + q - G) \delta(\omega \pm \omega_{q_j})$$

Structure factor    Population factor    Momentum    Energy

$$f_j^1(Q) = \sum_k b_k^{coh} \frac{Q \cdot \xi(q_j, k)}{\sqrt{m_k}} e^{-W_k(Q)} e^{iG \cdot r_k}$$

$$S_{inc}^1(Q, \omega) = \sum_k \frac{b_k^2}{\langle b^2 \rangle} e^{-2W_k(Q)} \frac{Q^2}{2m_k} \frac{g_k(\omega)}{\hbar\omega} \left[ n(\omega) + \frac{1}{2} \pm \frac{1}{2} \right]$$

$$g_k(\omega) = \int_j |\xi(q_j, k)|^2 \delta[\omega - \omega_j(q)] dq$$

$g^n(\omega) = A \frac{e^{-\hbar\omega/Q}}{Q} \left[ n(\omega) + \frac{1}{2} \pm \frac{1}{2} \right] S(Q, \omega)$

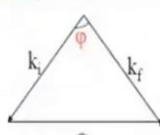
So, phonon density of states I mentioned you in the polycrystalline sample, but what we measure experimentally you can see dispersion relation is like this. But when we talk about phonon density of states, there is no  $q$  dependence, because as I told you, I have done measurements that many  $Q$  values, there is an averaging over all the phonon wave vectors ( $q$ )

because of the  $dQ$  values. And then when you add up we get this phonon density of states as a function of omega. I hope it is clear to you because there is a gap in  $\omega$  spectrum between the acoustic and optic modes. So, what we get, if I plot  $g(\omega)$  vs  $\omega$ , it looks somewhat like this. So, this is the gap, it is just shown in the perpendicular direction, but basically  $g(\omega)$  gives me density of states that means  $d\omega/dk$  for the entire sample, down there is a summation over all the phonon branches. So, it is only a function of  $\omega$  as I showed you earlier. And this is what we can measure, it is an easier measurement because summation of  $Q$  and  $q$  in a polycrystalline sample, the  $q$  is not defined, because all possible  $q$ 's are there, unlike a single crystal and there is nothing like orienting the crystal in a certain direction. So, I have got an average over all the  $q$ 's, when I do an experiment over large number of momentum vector transfers and I get neutron weighted dispersion relation which I mentioned here. So, how does it look like?

(Refer Slide Time: 28:08)

**Measurement of Phonon density of states**

**Vector diagram of scattering geometry**



$E_i(k_i)$  and  $Q$  are constant

$E_i(k_i)$  and  $\phi$  are changed such that  $Q$  is constant

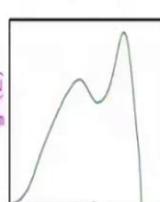
$$Q^2 = k_i^2 + k_f^2 - 2k_i k_f \cos\phi$$

$E_i$  and  $Q$  are constant in a single run

Obtain  $S(Q, E)$  at large number of  $Q$  values

Average all the data

Obtain  $g^{(a)}(E)$  from  $S(Q, E)$

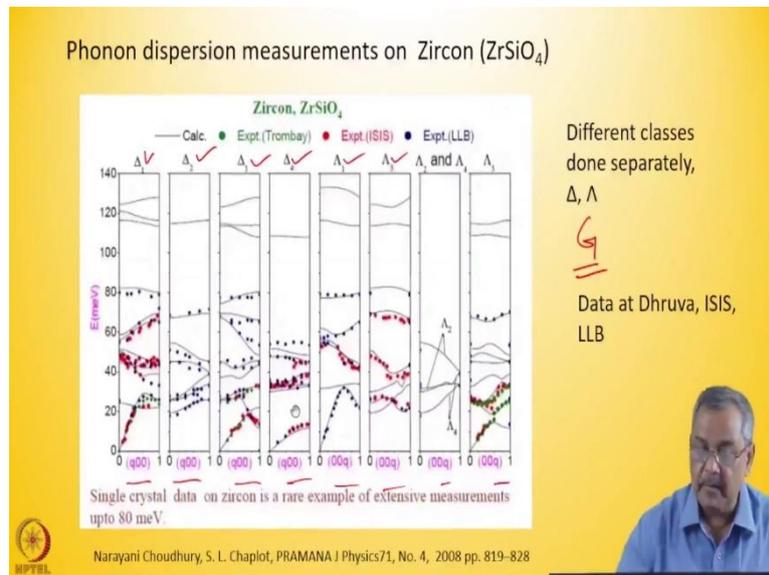
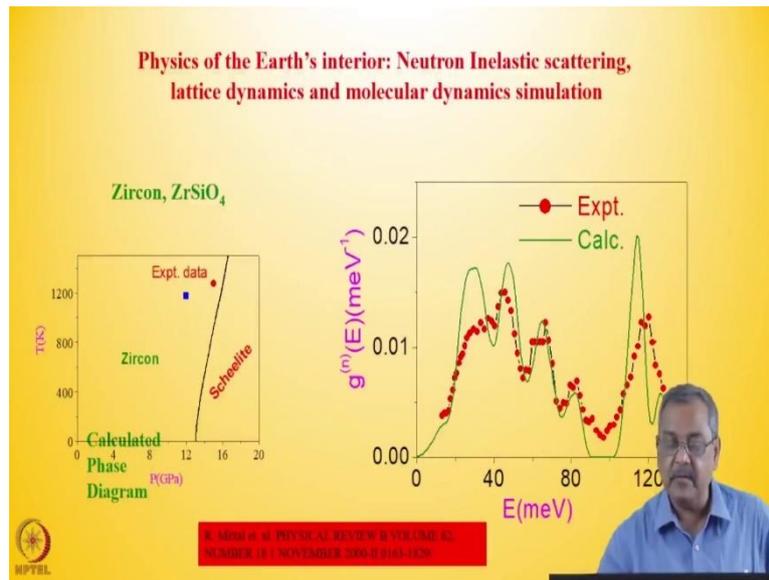
$$g^n(\omega) = B \sum_k \frac{4\pi b_k^2}{m_k} g_k(\omega)$$


$$g^n(\omega) = A \left\langle \frac{e^{-2W(Q)}}{Q^2} \frac{\hbar\omega}{n(\omega, T) + \frac{1}{2} \pm \frac{1}{2}} S(Q, \omega) \right\rangle$$

MPTCL

Well, this is what the experiment is, and I have shown you, how the data will look like.

(Refer Slide Time: 28:17)



Let me just show some data on dispersion relation measurement and density of states measurements using neutrons.

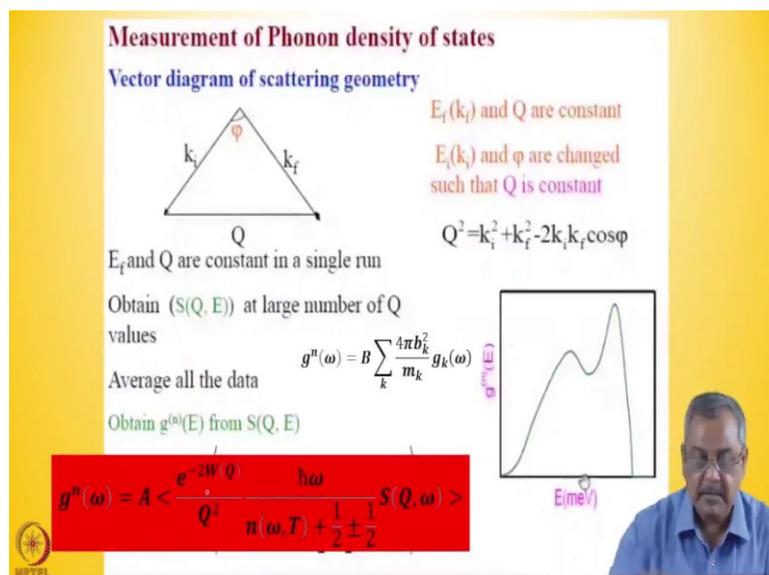
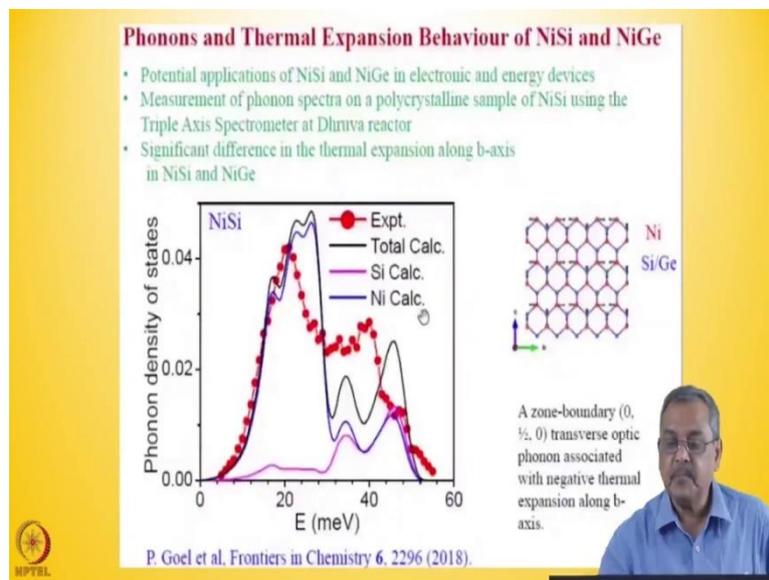
Earlier I had shown you this measurement on Zircon. This experiment also involves group theoretical calculations to find out which branches will be measured in an experiment and the corresponding  $G$  values. Earlier I listed out for all these measurements in  $(q00)$  direction for phonons. Various branches are measured in different experiments.

Now, for the same, when I do the density of states measurement, it is a much simpler experiment with powder samples and simpler data, you can see. And this is what we obtained as the fit between the experimental data (the red dots) and the calculated values. This feels

unlike fits to diffraction data, it is not a good fit. But this is considered a good fit for a density of states measurement. I mean, it is an ethical (or mathematical) question regarding how good a fit is, but in case of phonon density of states, this is considered a good fit.

If you do such an experiment, this fit will be considered good. And from here (density of states) I can get information about various macroscopic thermodynamic quantities like specific heat. This is one of the measurements done at Dhruva on density of states. Importantly, here we do not have  $q$  dependent data.  $q$  is averaged out. Experiment is done only as a function of energy transfer. So, it is easier to carry out and we get better neutron intensity.

(Refer Slide Time: 30:02)



$$S_{coh}^1(Q, \omega) = \sum_k \underbrace{\{f_j^1(Q)\}^2}_{\text{Structure factor}} \underbrace{\left[ n(\omega) + \frac{1}{2} \pm \frac{1}{2} \right]}_{\text{Population factor}} \frac{\hbar}{2\omega} \underbrace{\delta(Q + q - G)}_{\text{Momentum}} \underbrace{\delta(\omega \pm \omega_{qj})}_{\text{Energy}}$$

$$f_j^1(Q) = \sum_k b_k^{coh} \frac{Q \cdot \xi(q_j, k)}{\sqrt{m_k}} e^{-W_k(Q)} e^{iG \cdot r_k}$$

$$S_{inc}(Q, \omega) = \sum_k \frac{b_k^2}{\langle b^2 \rangle} e^{-2W_k(Q)} \frac{Q_l^2}{2m_k} \frac{g_k(\omega)}{\hbar\omega} \left[ n(\omega) + \frac{1}{2} \pm \frac{1}{2} \right]$$

$$g_k(\omega) = \int_j |\xi(q_j, k)|^2 \delta[\omega - \omega_j(q)] dq$$

These are examples of density of states measurements on NiSi and NiGe. Again, I am showing you the calculated value and the experimental data. This peak is not represented very well in the fit, but this peak is reasonably represented I would say. Here, one big problem is this, when I am writing  $g^n(\omega)$  here, you can see, the expression has  $e^{-2W_k(Q)}$ , that means, the  $k^{\text{th}}$  atom and its corresponding value of the Debye Waller factor at momentum transfer  $Q$ .

But here in this expression also you can see I am putting a Debye-Waller factor. When it is an average on  $Q^2$ , but  $W(Q)$  still remains. You can see that and this is neutron weighted density of state for all the atoms over here. The thing is that in a lattice, a heavier atom will have smaller amplitudes of motion compared to a lighter atom. So, this is an average  $W(Q)$  I am talking about. And this is an average, which I am using here, but actually it is not correct, because different atoms will have different amplitudes of movement. And when I am estimating or obtaining density of states, I am considering an average  $W(Q)$ . So, this is one source of error in our density of states calculation.

(Refer Slide Time: 31:56)

Handwritten notes and a video frame. The notes include:

- $\underline{Q}$  (underlined)
- $Q$
- $\underline{Q}$  is large?
- $Q$
- $g(\omega)$
- $0$
- $e^{-w(Q)}$

The video frame shows a man in a blue shirt speaking.

Handwritten notes and a video frame. The notes include:

- $\underline{Q + \bar{q} = \bar{G}}$
- $Q + dq \sim \text{few}$
- Reciprocal
- $\{q_i\}$
- incoherent approximation to coherent phonon

The video frame shows a man in a blue shirt speaking.

Also, as I told you earlier, that the momentum transfer  $Q$  is chosen large because then the uncertainty in  $dQ$  adds up over many phonon  $q$  values as I showed you in the drawing here. But the fact is that if I go to very large  $Q$ 's, then again, my Debye Waller factor will kill the intensity because Debye Waller factor shows that the intensity decreases with  $Q$ .

So, again I have to take an optimal  $Q$ . It should be large enough to work with incoherent approximation, but not so large that Debye Waller factor kills my neutron intensity. So, these are the things which have to be taken care of and this is the kind of data that we get experimentally. I think here I will call it a stop for discussion on phonons.

We have discussed the phonon dispersion relations using single crystals and their measurements and phonon density of states that we can measure using polycrystalline samples which are easier experiments. I stop here the discussion on phonons, the collective motion of atoms in a crystalline solid, and in the next part, I will go on to quasi-elastic neutron scattering for stochastic motions.