Keywords: Pair Correlation function, Scattering law, Structure, dynamics

In this lecture, I will again go back to the correlation function G(r,t) whose double Fourier transform over time and momentum gives me the S(Q,ω), the scattering law that I tend to measure in an experiment. I will try to familiarize you with G(r,t) and its physical significance. As S(Q,ω) and G(r,t) are Fourier transform of each other, I will also introduce you to the design requirements of spectrometers depending on the kind of dynamics or kind of structure that you want to see. You need to decide it beforehand and that will dictate how the detectors are positioned, how energies are measured and everything.

I will take you through a tour in various major neutron sources and our own source and show various components that are present in a neutron scattering setup. Starting right from the source to the detector, I will take up all these descriptions a little briefly to introduce you to the proper experimental facilities.

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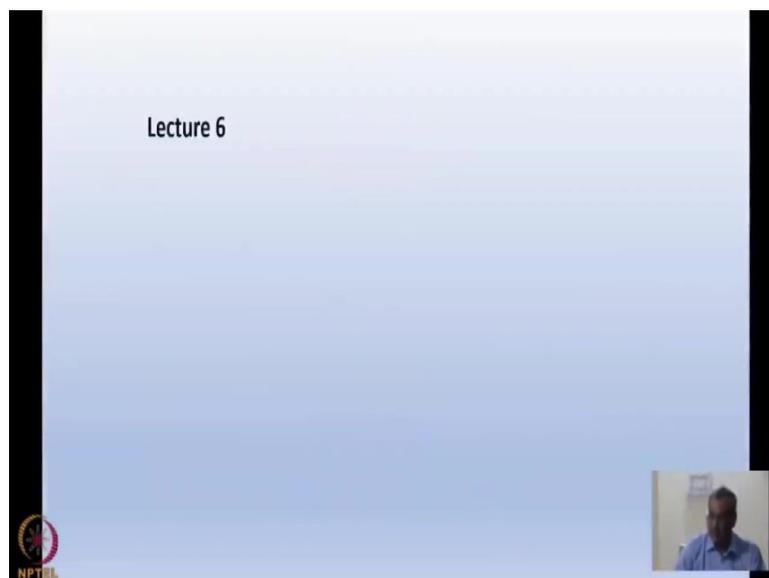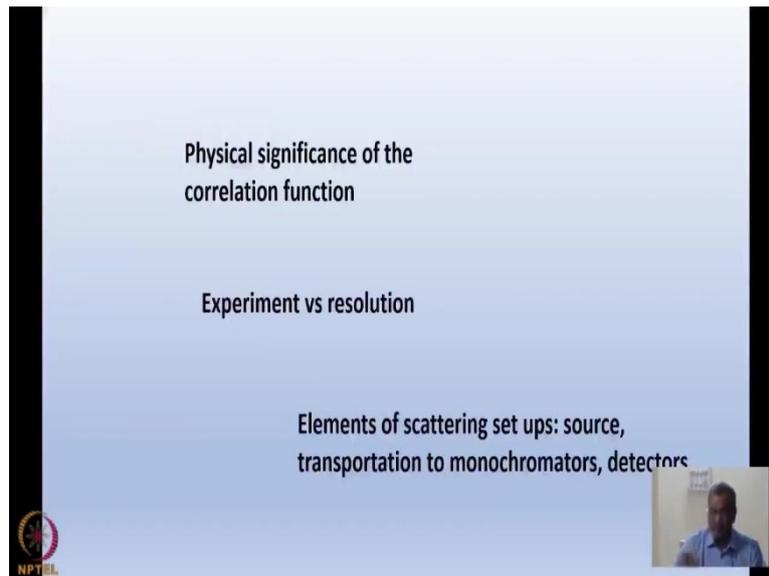
In this lecture, I will discuss the physical significance of the correlation function, I will also discuss the design of experiments versus resolution and then I will introduce you to elements of scattering setups; the source, beam transportation, beam monochromatization to neutron detectors, I will touch upon all of them briefly if not in this lecture, then in the next lecture.

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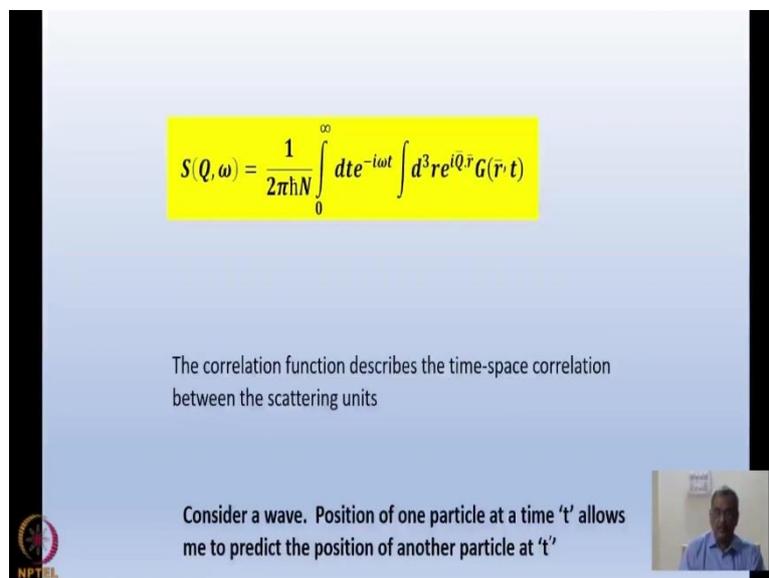
I had indicated that $S(Q,\omega)$ is a double Fourier transform of $G(r,t)$ where integration over d^3r (space) with $e^{iQ.r}$ takes me to the Q-space and integration over dt (time) with $e^{i\omega t}$ takes me to the omega space. I need instruments in which specifically I measure neutrons at a certain angle. And if there is an attempt to understand dynamics, then we also try to find out what is the energy of the scattered neutron. The dynamical process in a system like phonons, diffusion, molecular vibrations, they can exchange energy with the neutrons. Why it is not possible with

the x-ray is due to the fact that 1Å X-ray is equivalent to about 10 keV energy. Hence to measure the change of meV on a keV x-ray is difficult. Nowadays, in synchrotron sources using very high resolution back scattering instruments, one can measure energy transfers even of that order. But in general, I can broadly tell you that to measure the energy transfer of meV using a photon of keV is like trying to measure a milli Volt change in voltage using a voltmeter which is calibrated for Mega voltage measurements.

Now, let us come to the correlation function, which describes a time-space correlation between the scattering units. Consider a wave. The position of one particular at time t allows me to predict the position of another particular at time t'.

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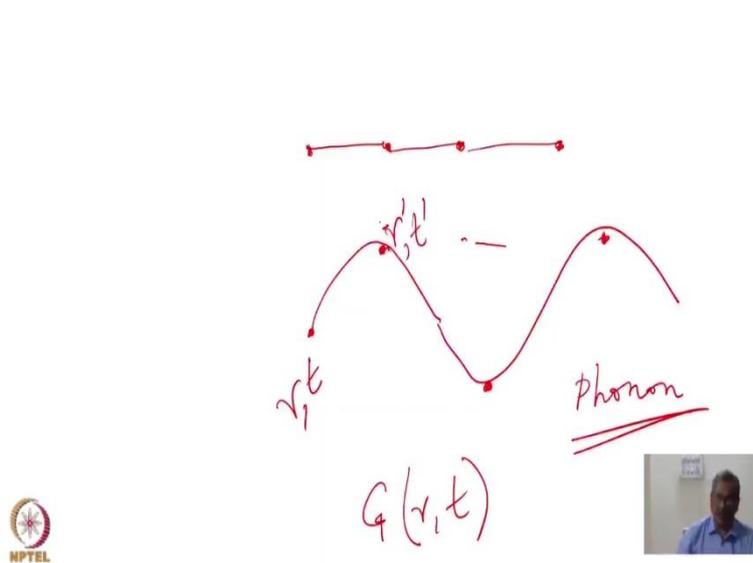
Let us consider in a linear chain of atoms in a system. I know that if there is a phonon then the phonon possibly travels like this; that means the displacement of this particle/atom is correlated to the displacement of that particle. It is also correlated to the displacement of this particle and this particle etc. So, at any instant t, if I know the position of this particle r that is (r,t), then the position of all other particles at any other position r' at a time t' is known for a phonon. That means, this phonon dispersion relation gives me $G(r,t)$ for this system.

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Now coming back to a static system, if I consider classical G(r,t) given by,

$$G^{cl}(r,t) = \sum_j \langle \delta[r - \{\bar{R}_0 - \bar{R}_j(t)\}]\rangle$$

At $t = 0$, it can be written as,

$$G(r,t) = \delta(r) + \sum_{j \neq 0} \langle \delta[r + R_j - R_0]\rangle = \delta(r) + g(r)$$

These are sum of delta functions. I sum over all the partners and do an ensemble averaging. The ensemble average is inherent, because when we do a measurement over finite time, we are actually taking frame after frame after frame on to the detector and that does the ensemble averaging automatically. This ensemble average is shown by the angular brackets here.

At $t = 0$, this has two parts. A particle with its own location, $\delta(r)$. Then for all other partners of it, delta function peaks whenever $R_j - R_0$ hits another atom, g(r).

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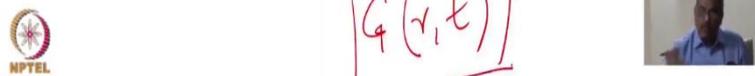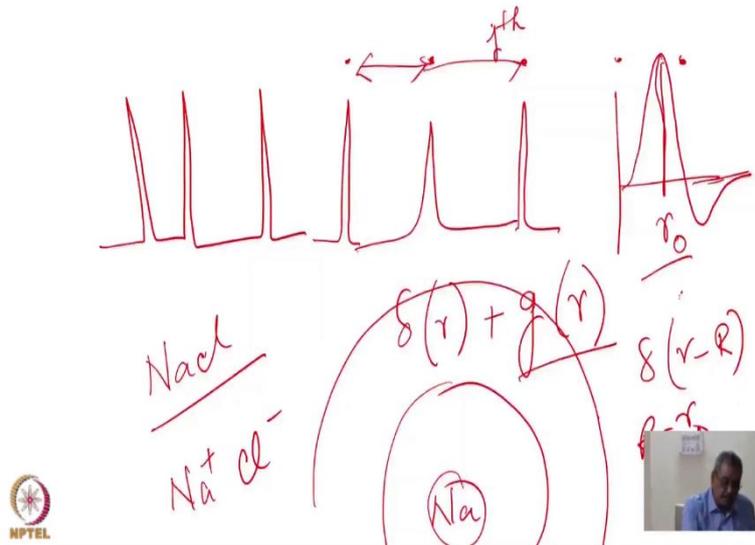
Let us again consider the static chain of atoms. It has got a delta function at the origin and any particle can be at the origin. So, it is a $\delta(r)$ for this for all the particles means that whenever the distance is such that there is a particle there, I have got a delta function peak. Along with this, there is also a pair correlation function $g(r)$. What do you mean by a pair correlation function?

As a simple example, suppose I take a case of molten sodium chloride. At any instant of time there are Na^+ and Cl^-. At an instant of time, if I look at this molten salt and take a Na ion then statistically it is surrounded by a circle of chlorine ions because they are ions and they attract each other. Then statistically, this circle of chlorine ions is surrounded by an outer circle of sodium ions. Inner shell compared to outer shell is more clearly defined as you go farther and farther. These shells that I am showing you, they get more and more diffused and finally I have constant density. So, starting with one sodium as origin, I have got a pair correlation function,

which will possibly look something like this. After delta function peak, there is pair correlation peak, which gets smaller and smaller with distance.

In the first shell, shell, I have got a peak, say at r_0, means for $\delta(r\text{-}R)$ delta function peak is at R = r_0. Later we will discuss that it comes out naturally for liquid and amorphous systems. What I mean is that, in a static system, in a crystalline system, you have got sharp delta function peaks, but it does not mean that when we have a liquid or amorphous system, there are no peaks: they are diffuse, but there is very clear short-range order in such systems and that can be found out from the g(r) which we can measure in an experiment. In g(r) we have taken out time and for such experiments we are looking at the structure but not the dynamics in the system. This was a brief introduction to g(r) or pair correlation function for a static case.

For a dynamic case given a phonon, say transverse acoustic phonon, the displacement of all the particles are correlated, and their g(r,t) is also well known. So, you can find out S(Q,ω) for the dynamic case also. With this I have introduced you to g(r,t) for a static case and for a dynamic case.

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$$S(Q,\omega) \sim \int e^{i(Q.r - \omega t)} G(r,t) d^3 r dt$$

Since S(Q,ω) and G(r,t) are Fourier inverse of each other, let us invoke "Uncertainty" principle for designing an experiment

$$\Delta P \Delta r \sim \hbar; \quad \Delta E \Delta t \sim \hbar$$

The uncertainty 'ΔK' comes from the range of momentum transfer in the scattering experiment

The uncertainty 'ΔE' comes from the range of energy transfer

You need to choose the experiment depending on what you want to see!!

Now, I will come to resolution in an experiment and what can I do about it. We know that Q or $\hbar Q$ is the momentum transfer. Hence, Q represents the momentum space and r is the real space. And, they are related by the uncertainty relation, $\Delta P \Delta r \sim \hbar$, $\Delta P \Delta r$ should be greater than or equal to $\hbar$. Similarly, the energy transfer is $\hbar\omega$ and energy and time are also in uncertainty relation because they are Fourier inverse of each other. So, $\Delta E \Delta t$ should be greater than or equal to $\hbar$.

Now, in an experiment, the ΔQ is basically the range of momentum transfer that we measure in the experiment. It gives me the limit of uncertainty for a given system. It also tells me that how much is a quantum mechanical space resolution that I can achieve. So, we need to design or choose to experiment depending on what you want to see.

Let us consider a diffraction experiment, quantum mechanics says the spatial resolution in an experiment is given by, $\Delta r = \frac{2\pi}{Q_{max}}$. 0 to Q_{max} is measurement range. We know $Q = \frac{4\pi}{\lambda}\sin\theta$ when the beam is deviated by 2θ.

Let k_1 be the incoming wavevector and k_2 be the outgoing wavevector then $Q = k_2 - k_1$ and $Q = \frac{4\pi}{\lambda}\sin\theta$. So, $Q_{max} \propto \theta_{max}$, θ_{max} is how much theta I am covering in an experiment. Here I assumed that λ is constant, means it is a constant wavelength experiment. From the expression of Q, we can see that Q can also be changed by changing the wavelength, it will become larger if wavelength (λ) is reduced. This is what is done in spallation neutron sources, but that will come later.

What I mean is that, here, in an angle dispersive experiment, Q_{max} depends on the maximum angle of the experiment.

Let us say I want to go for a wavevector transfer of 10 Å⁻¹ with 1.2 Å incident neutrons. For this need to go to scattering angles of around 140° that is $2\theta = 140°$. In a typical powder diffractometer, we have 10 to 12 Å⁻¹ range. So, we had to have detector moving from 0 to 140°. And this gives me a quantum resolution of 0.6 Å for measurement of the lattice parameters, and that is reasonable, because often lattice parameters are in the range of this.

Now, suppose I want to broaden the resolution. That means, I do not want to see the atomic structure, I want to see some larger conglomerations. Say, I make my resolution 30 Å. This is not improving the resolution but it is making the resolution poorer. And, I am using let us say a 4 Å neutron. We are using 4 Å neutron because with a 4 Å neutron for having a resolution of 30 Å we need to go to about 4°. If I use shorter wavelength neutrons shorter wavelength neutrons then to go to these kinds of smaller Q values, theta also has to be much smaller. Now you have a direct beam and you have to measure up to 4°, which is a reasonably small angle. If we have to go to 1°, because I want to go to 0.2 Å⁻¹, using a shorter wavelength neutron, then, such small angles are difficult because you also have a direct beam which has got a beam width of certain angular spread. And the direct beam might start getting into this. So, that is why I said that I will be using 4 Å neutrons, which is called a cold neutron. I will tell you how we get cold neutrons.

This one the top one in the yellow rectangle, this is a typical value for a diffractometer for crystallographic structure, where we are going up to a wavevector transfer of 10 Å⁻¹. Similarly, for a resolution of 30 Å, the other way around, we may need to go to 0.2 Å⁻¹. Look at the difference here from 10 Å⁻¹ to 0.2 Å⁻¹. To have 0.2 Å⁻¹ with a 4 Å neutron we need to go to an angle of around 4°. This is a typical example of small angle neutron scattering (SANS). Here we do not see the medium as consisting of atoms and crystallographic lattice, but we are looking at conglomerations which may be 30 Å large. Sometimes, it can be even larger, 100 Å, 200 Å. These are inhomogeneities in the system that we may be studying. So, the need of what do we want to see, dictates the spectrometer and we have to design the spectrometer depending on this need.

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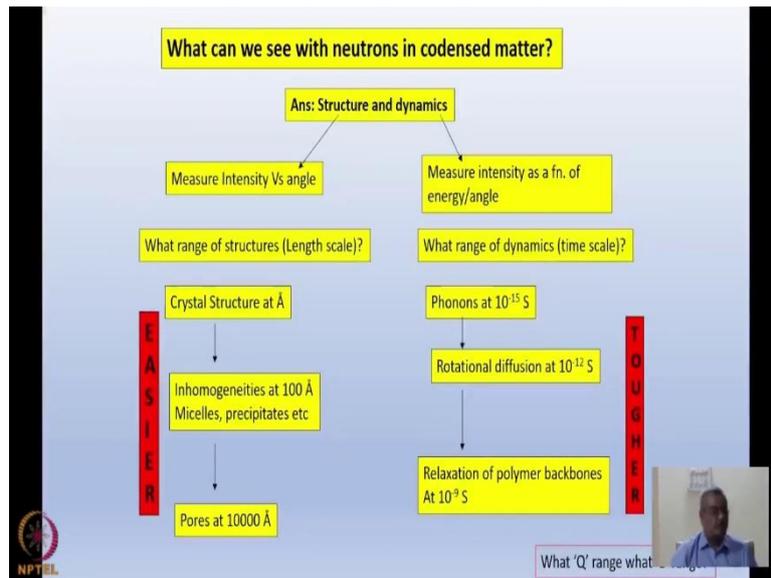
I will come back to this in the next part of the talk, the what we can see with neutrons in condensed matter.