Fundamentals and Applications of Dielectric Ceramics

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Lecture No. – 22

Frequency Dependence of Dielectric Constant

Welcome again to the new lecture of this course, fundamentals and applications of dielectric ceramics.

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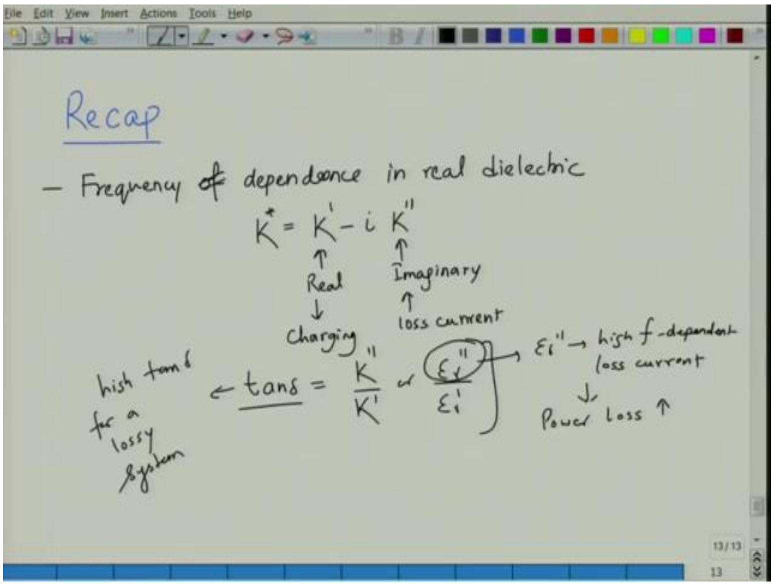

Recap

- Frequency of dependence in real dielectric

$K^* = K' - i K''$

Real ↓ Charging

Imaginary ↑ loss current

high tanδ for a lossy system ← tanδ = $\frac{K''}{K'}$ or $\frac{\varepsilon_i''}{\varepsilon_i'}$

ε_i'' → high f-dependent loss current ↓ Power loss ↑

Let me briefly recap the previous lecture. In the previous lecture we talked about frequency dependence in real dielectrics and there we defined dielectric properties in terms of complex behavior. So, we defined the k* as:

$$k^* = k' - ik''$$

So, the first term in L.H.S is the real part and the second is the imaginary part. So, basically, this real part represents the charging current and this imaginary part represents the loss current.

Basically these are frequency dependent components. In addition, you will have frequency independent components which is separate. And the ratio of these tan δ :

$$\tan\delta = \frac{k''}{k'}$$

So, the ratio of these two is tan δ. So, for a lossy system we will have high tan δ.

Basically what it means is that if your system has high ε'' which means it has high frequency dependent loss current. So, high ε_r'' will mean high frequency dependent loss current and what this would also mean is that your power loss will also increase. So, for dielectric materials you have ε_r'' which represents the actual dielectric behavior, whereas ε_r'' represents the loss behavior of a dielectric. So, these are taking the frequency dependent parts into consideration.

On top, you might also have ohmic losses. So, the total losses have to be calculated by combining ohmic losses plus the non-ohmic frequency dependent losses. So, this is what we have done in the last class and we determined the framework to do that.

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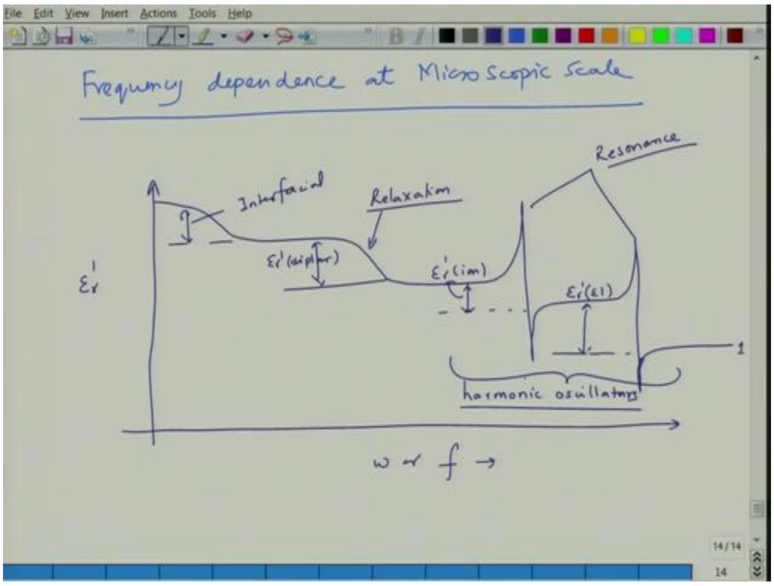

Frequency dependence at Microscopic Scale

Now, what we will look at is the frequency dependence at the microscopic scale. So, we saw earlier that your dielectric constant varies like this. So, you have frequency, you can write ω or f, then we have ε_r. Now that we know it is real and imaginary part we will plot the real part. This is what we plot here. So, initially we just plot the scalar part but here we know that it is a real part. So this varies in this fashion.

So, at high frequencies your dielectric constant is one, at little bit lower frequency your dielectric constant is ε_r' **(electronic)**. At lower frequency than that , it is ε_r' **(ionic)**. And here anything on top of one is this, and this becomes ε_r' **(dipolar)**. And then we have interfacial. That is what we said earlier. Now, we see some interesting features here.

The interesting features are that at high frequencies when you transition from one to ε_r' **(electronic)** there is a resonance kind of peak. What is the reason of this resonance? Similarly, as you transition from electronic to ionic you see again a similar resonance kind of peak. These are the peaks also encountered in mechanical oscillators. When you have a resonance you have these kinds of resonance peaks.

So, we will model this ionic and electronic polarization on the basis of basically harmonic oscillators. So, these are two, we will call them as resonance peaks, and we will see how they come about. And then, when you go to lower frequencies you do not see resonance peaks as well, but what you see is a slow decay of dielectric constant. This is called as a relaxation because here at the molecular level there are statistical events in response to time.

Basically, when you have electric field applied and when you have certain frequency of it, the dipoles as you go from lower to higher frequency or higher to lower frequencies they take time to align themselves in the direction of applied field. This is because the dipoles are heavier. They have certain, you can say, if you like you can call it friction in the lattice and they have to go from one stable state to another stable states.

As a result, there is energy barrier to be crossed. So, as a result, they slowly relax from one position to another position and that is why it is a time taking process and this process is called as relaxation. So, we will look at these three phenomena. We will not go into details of interfacial. We will look at mainly dipolar relaxation and resonances related to ionic and electronic polarisation considering simple harmonic oscillator model.

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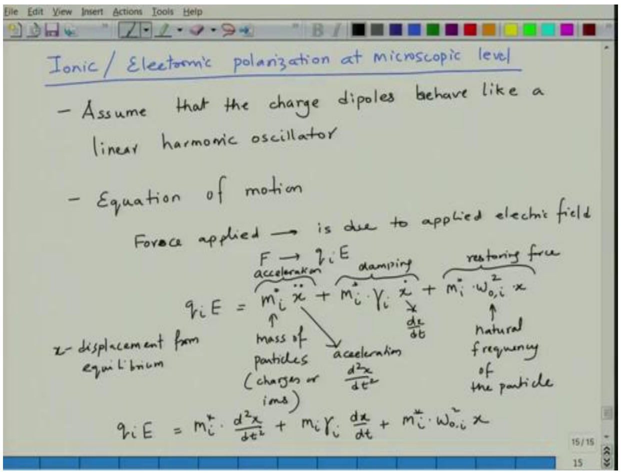

Ionic / Electronic polarization at microscopic level

- Assume that the charge dipoles behave like a linear harmonic oscillator
- Equation of motion

Force applied → is due to applied electric field

F → $q_i E$

$$q_i E = \overbrace{m_i^* \ddot{x}}^{\text{acceleration}} + \overbrace{m_i^* \cdot \gamma_i \cdot \dot{x}}^{\text{damping}} + \overbrace{m_i^* \cdot \omega_{o,i}^2 \cdot x}^{\text{restoring force}}$$

x - displacement from equilibrium

mass of particles (charges or ions)

acceleration $\frac{d^2x}{dt^2}$

$\frac{dx}{dt}$

natural frequency of the particle

$$q_i E = m_i^* \cdot \frac{d^2x}{dt^2} + m_i \gamma_i \frac{dx}{dt} + m_i^* \cdot \omega_{o,i}^2 x$$

So, let us do that. This is basically the first thing, it is the ionic and electronic polarisation at microscopic level. First what we do is we assume that the charged dipoles behave like a linear harmonic oscillator. This is the assumption that we make to begin with. So, if they do that, then they also follow the equation of motion. So, now we have to write what is the equation of motion in response to a force:

$$q_i E = m_i^* \ddot{x} + m_i^* \gamma_i \dot{x} + m_i^* \omega_{o,i}^2 x$$

So, you have applied force. As a result, they will have certain acceleration, they will have a damping component and so on and so forth. So, let us say, in this case, the force applied basically you can say is due to applied electric field which is equal to, let us say q_iE. This F is nothing but the q_iE. So, q_iE is the force applied. This is equal to the mass of entity, that is, m_i.

It could be electrons with respect to the nucleus or it could be ions in the case of ionic polarization. So, this is basically mass of the particle you can say, it could be charges, or ions, all right? Mass multiplied by acceleration which is $\ddot{x}$. Then we write m_i^* into γ_i into $\dot{x}$. And what is this x? Here, we write x as a displacement from equilibrium.

So, $\ddot{x}$ mean basically acceleration or you can say this is:

$$d^2x/dt^2 = \ddot{x}$$

The second thing that will happen is every system harmony accelerator will feel friction. As a result, there will be a friction coefficient γ_i and this is nothing but γ_i into m_i multiplied by velocity which is dx /dt. This is from classical mechanics. And then, we have another term which is essentially m_i^* multiplied with $\omega^2_{0,i}.x$.

And the third term is basically if it is a simple harmonic oscillator where the charges are connected through a spring, there will be a restoring force, all right? So, there will be acceleration, there will be restoring force, and there will be damping. So, the first term basically is due to, you can say this term is because of acceleration. The second term is because of, you can say damping or frictional force.

And the third term is because of restoring force. So, essentially, this is nothing but **k.x**. So, we have converted this into **mω^2x**. So, here $\omega_{o,i}$ is the natural frequency of the particle. So, we can say $\omega_{0,i}$ or $\omega_{i,0}$ is natural frequency of the particle i, and x is the displacement. So, we can write this equation. Alternatively you can write this as:

$$q_i E = m_i^* \frac{d^2x}{dt^2} + m_i^* \gamma_i \frac{dx}{dt} + m_i^* \omega_{o,i}^2 x$$

This is the equation of motion which will be in this. So, you will have applied force. In response to apply force you will have acceleration, every particle will feel acceleration, and then you will have damping, and you will have a restoring force because of the spring through which it is connected to which we assume. So, this field is basically the field that we actually apply, that the molecule will feel, or the system will feel.

It will be a local field that is there at present, and the local field in dielectrics could be slightly different as compared to the applied field, but for the sake of simplicity we will take it as applied field itself. So, we can assume that you have a system, just like a gas in which there are n atoms per unit volume and you consider them to be non interacting. So, if the dipoles were interacting, then the field would be different, but we assume that dipoles are not interacting, and as a result, the field is same as applied field.

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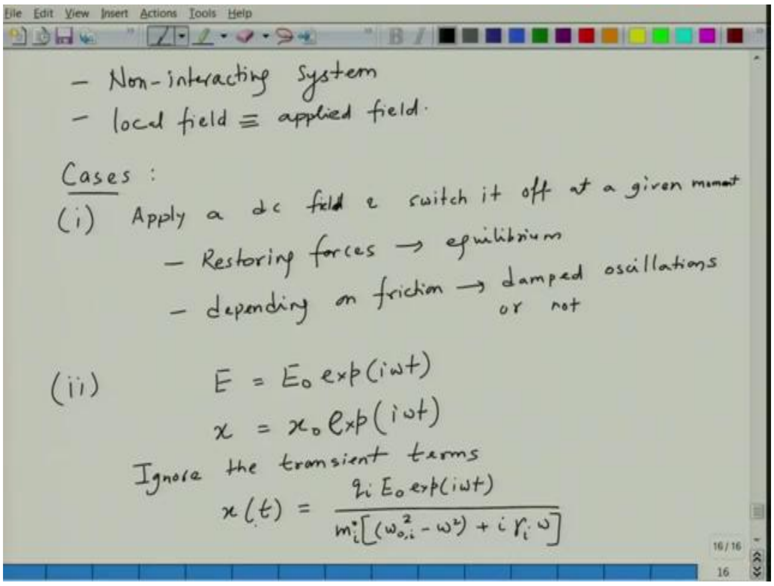

- Non-interacting System
- local field $\equiv$ applied field.

Cases :

(i) Apply a dc field & switch it off at a given moment

- Restoring forces → equilibrium
- depending on friction → damped oscillations or not

(ii) $E = E_0 \exp(i\omega t)$

$x = x_0 \exp(i\omega t)$

Ignore the transient terms

$$x(t) = \frac{q_i E_0 \exp(i\omega t)}{m_i^*[(\omega_{o,i}^2 - \omega^2) + i\gamma_i \omega]}$$

So, we can say that system is non interacting system. So, as a result, we will have local field which is same as applied field. So, now, you consider two cases. The first case is you apply a dc field and then switch it off at a given moment. What will happen when you switch off the electric field which is d.c, the restoring forces will pull back the charges to equilibrium. So, basically you will have restoring forces.

They will establish equilibrium by pulling the charges back to their original positions and if there is no friction in the system and there is no damping, as a result, there will be no damping of oscillations. And if you have friction in the system, you will have damped oscillations. So, depending upon friction we will have damped oscillations or not. This is the case of dc field which is not very interesting because we are not interested in dc field and we know that in dc field there is no frequency dependence there.

So, we will have something like ohmic losses in the system. And now, the second case that is there is when field is like this. So, when field E:

$$E = E_0 e^{i\omega t}$$

It is sinusoidally varying field. So, if you have field which is sinusoidally varying, then it is likely that the displacement will also vary sinusoidally. So, the x displacement is also expressed as:

$$x = x_0 e^{i\omega t}$$

So, if you ignore the transient terms, students who are more interested they can go through the solution by themselves by substituting x = $x_0$exp(iωt), but for the sake of simplification, for the sake of saving time, we just write the solution as:

$$x(t) = \frac{q_i E_0 e^{i\omega t}}{m_i^* [(\omega_{0,i}^2 - \omega^2) + i\gamma_i \omega]}$$

This will be the solution of the equation that we have just written in the previous pages. Assuming that field is sinusoidally varying, if the field is sinusoidally varying, it is likely that the response will also be similar. Responses is the displacement. So, displacement is also x is equal to exp(iωt). If it has this form, then you substitute this in the above equation, solve the equation, ignore the transient terms, and you will get this kind of solution for x.

Now, x will vary as shown in the equation above. This will what you will observe.

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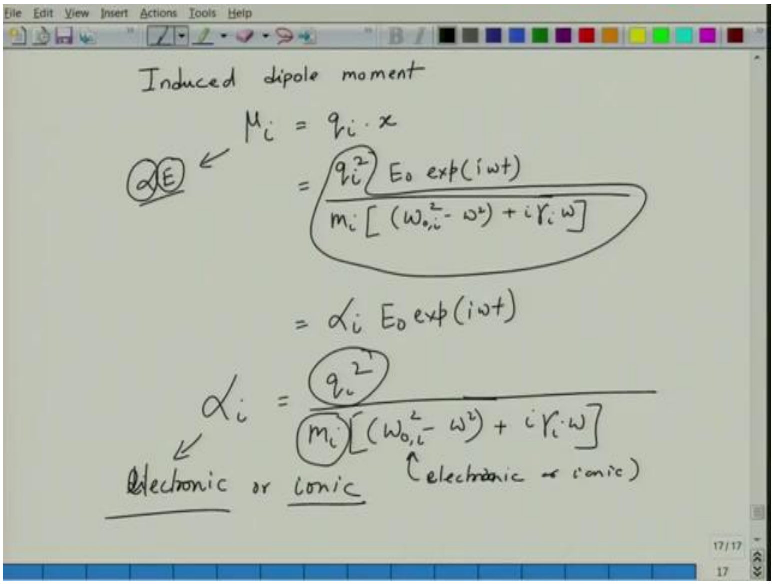

Induced dipole moment

$$\mu_i = q_i \cdot x$$

$$\alpha E \leftarrow$$

$$= \frac{q_i^2 E_0 \exp(i\omega t)}{m_i[(\omega_{0,i}^2 - \omega^2) + i\gamma_i \omega]}$$

$$= \alpha_i E_0 \exp(i\omega t)$$

$$\alpha_i = \frac{q_i^2}{m_i[(\omega_{0,i}^2 - \omega^2) + i\gamma_i \cdot \omega]}$$

Electronic or Ionic

(electronic or ionic)

Now, what is the induced dipole moment if this is the case. The induced dipole moment is equal to:

$$\mu_i = q_i . x = \frac{q_i^2 E_0 e^{i\omega t}}{m_i^*[(\omega_{0,i}^2 - \omega^2) + i\gamma_i \omega]}$$

and what is μ? We know that:

$$\mu = \alpha . E$$

That is the polarisability multiplied by electric field. This can be also written as:

$$\mu_i = \alpha_i E_0 e^{i\omega t}$$

So, the α term what we have just derived here is basically the polarisability. So, this is α_i of an i kind of species is equal:

$$\alpha_i = \frac{q_i^2}{m_i^*[(\omega_{0,i}^2 - \omega^2) + i\gamma_i \omega]}$$

This could be electronic or ionic. The change will be in the charges, the charges will be different for electronic system and the ionic system.

The mass will be higher for ionic system than for electronic system. As a result, the polarisability of ionic systems will be lower as compared to that of electronic systems. What you also have here is you have the term $(\omega^2_0 - \omega^2)$ when $\omega = \omega_i$ for this corresponding. So, this $\omega_{0,i}$ will mean you will have either electronic or ionic.

So, this basically, you can say, is the characteristic frequency at which you will have anomaly here. The term will go to infinity. If you ignore this term, the right term, this will go to infinity, and this is where the resonance will occur. So, when your omega is equal to $\omega_{0,i}$ for electronic and for ionic polarization, that is where you will have resonance.

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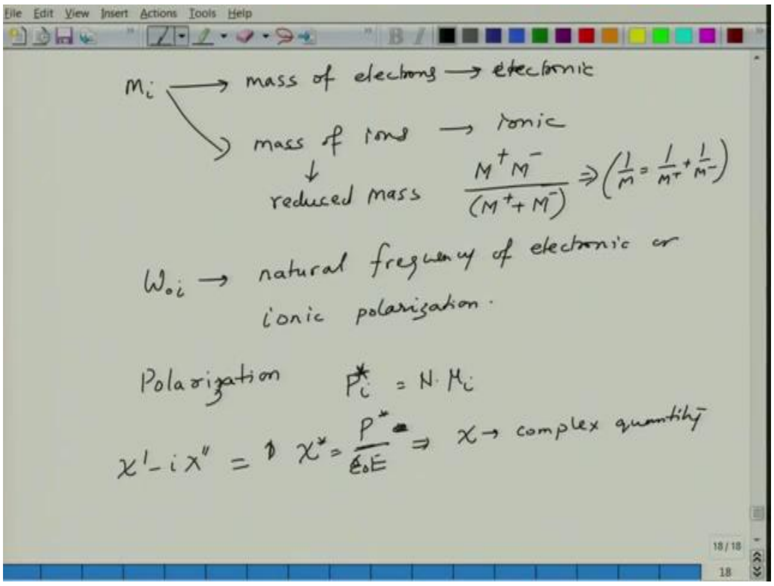

$m_i \rightarrow$ mass of electrons $\rightarrow$ electronic

$\searrow$ mass of ions $\rightarrow$ ionic

$\downarrow$ reduced mass $\frac{M^+M^-}{(M^+ + M^-)} \Rightarrow \left(\frac{1}{M} = \frac{1}{M^+} + \frac{1}{M^-}\right)$

$W_{oi} \rightarrow$ natural frequency of electronic or ionic polarization.

Polarization $P_i^* = N \cdot \mu_i$

$\chi' - i\chi'' = \chi^* = \frac{P^*}{\epsilon_0 E} \Rightarrow \chi \rightarrow$ complex quantity

So, here you can consider m_i could be either mass of, you can say, electrons. This will be for electronic polarization, right, or it could be mass of ions for ionic polarisation system. Generally for ionic polarisation we consider what we say is reduced mass of the system which is given as:

$$M = \frac{M^+M^-}{M^+ + M^-} or \frac{1}{M} = \frac{1}{M^+} + \frac{1}{M^-}$$

So, M^+ and M^- are cationic and anionic masses. Similarly, q_i will be as appropriate, that can be easily determined, and the $\omega_{0,i}$ will be natural frequency of electronic or ionic polarisation. And this is why you see when you plot them. The plots will come later on. So, let us write now the polarisation, what is the polarisation going to be? The polarisation is nothing but:

$$P_i^* = N.\mu_i$$

We know that we can just multiply the number of dipoles. Now, we know that, since here we have complex terms, we know that susceptibility was equal to:

$$\chi^* = \frac{P^*}{\varepsilon_0 E} = \chi' - i\chi''$$

Now, the susceptibility also becomes a complex quantity because your μ is complex quantity, as a result, your P is complex quantity, as a result, your susceptibility will also be complex quantity.

If you now segregate the terms, that is, we used what we call as equations, let just see, let us go back to some previous equations, so, basically similar equations. Similarly, you will have to resolve the P into real numbers and complex numbers.

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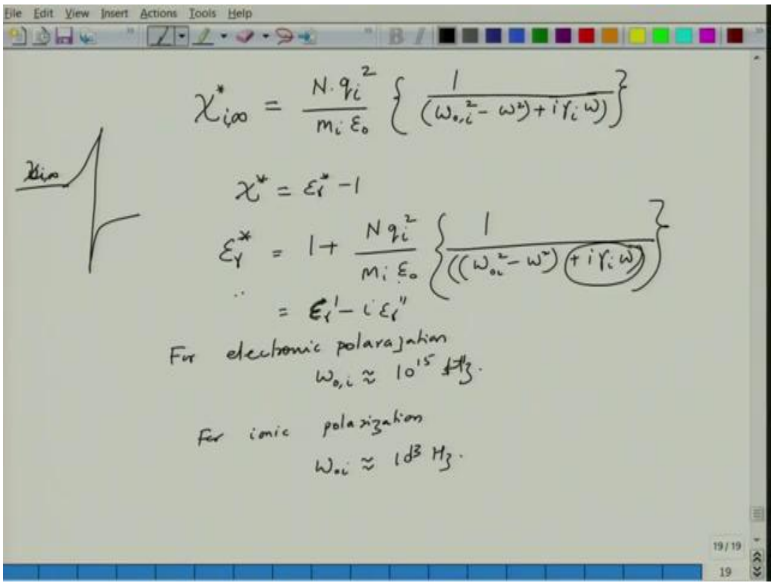

$$\chi^*_{i\infty} = \frac{N \cdot q_i^2}{m_i \varepsilon_0} \left\{ \frac{1}{(\omega_{0,i}^2 - \omega^2) + i\gamma_i \omega)} \right\}$$

$$\chi^* = \varepsilon_r^* - 1$$

$$\varepsilon_r^* = 1 + \frac{N q_i^2}{m_i \varepsilon_0} \left\{ \frac{1}{((\omega_{0i}^2 - \omega^2) \boxed{+ i\gamma_i \omega})} \right\}$$

$$= \varepsilon_r' - i\varepsilon_r''$$

For electronic polarajation

$\omega_{0,i} \approx 10^{15}$ Hz.

For ionic polarization

$\omega_{0i} \approx 10^{13}$ Hz.

So, you do the resolution that you can do yourself, that is a simple exercise. After resolving the real and imaginary, first we get the complex quantity itself. The complex quantity is $\chi^*_{i,\infty}$ .Star means it is a complex quantity first of all. So, that is why we write polarisation as a complex quantity. Infinite is a term which is written basically. These are the susceptibilities or dielectric constants below natural frequencies.

Since we will see we will have a resonance like this, so when you measure them below the natural frequency it means it is susceptibility i infinity. So, that is a nomenclature thing. So, basically $\chi^*_{i,\infty}$ can be written as:

$$\chi^*_{i,\infty} = \frac{N.q_i^2}{m_i\varepsilon_0}\left\{\frac{1}{(\omega_{0,i}^2 - \omega^2) + i\gamma_i\omega}\right\}$$

It is nothing but P/ε_0E.

The E and E will cancel each other. As a result, we will have this term. And from this we can determine ε_r. We know that:

$$\chi^* = \varepsilon_r^* - 1$$

So, ε^*_r will be equal to:

$$\varepsilon_r^* = 1 + \frac{N.q_i^2}{m_i\varepsilon_0}\left\{\frac{1}{(\omega_{0,i}^2 - \omega^2) + i\gamma_i\omega}\right\}$$

As it turns out, for electric polarisation, $\omega_{0,i}$ is of the order of 10^{15} s^{-1} or you can say Hz.

And for ionic polarisation, these are approximate numbers, $\omega_{0,i}$ is approximately 10^{13} s^{-1} or Hz. And you can see that for the calculation of susceptibility and dielectric constant you have to use the frequencies which are lower than the natural frequency for electronic polarisation assuming that the damping term will be very small, negligible. So, from this you can also determine what is ε_r.

So, that is equal to:

$$\varepsilon = \varepsilon_r' - i\varepsilon_r''$$

Now, you have an **i** term here. So, what you will do is that you will separate these variables into real and complex parts.

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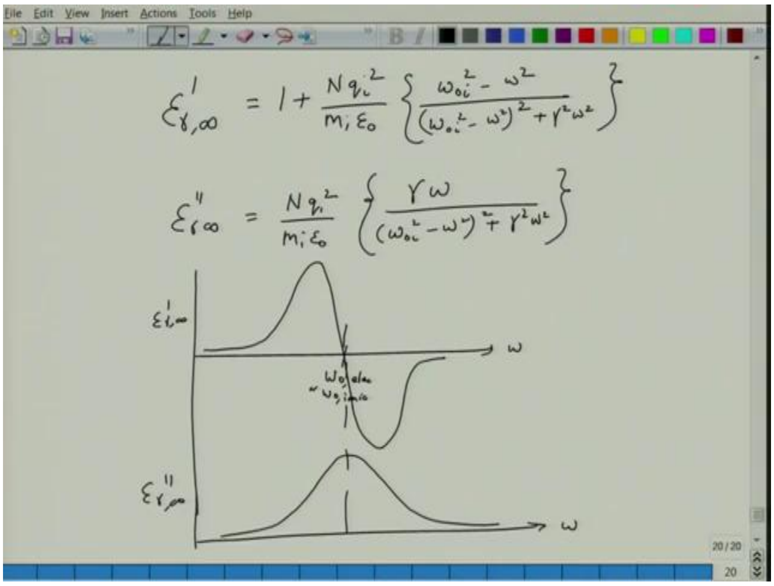

$$\varepsilon'_{r,\infty} = 1 + \frac{Nq_i^2}{m_i\varepsilon_0}\left\{\frac{\omega_{0i}^2 - \omega^2}{(\omega_{0i}^2 - \omega^2)^2 + \gamma^2\omega^2}\right\}$$

$$\varepsilon''_{r\infty} = \frac{Nq_i^2}{m_i\varepsilon_0}\left\{\frac{\gamma\omega}{(\omega_{0i}^2 - \omega^2)^2 + \gamma^2\omega^2}\right\}$$

When you do the separation into real and complex parts you will find $\varepsilon_{r,\infty}$ for ionic and electronic as:

$$\varepsilon'_{r,\infty} = 1 + \frac{N.q_i^2}{m_i\varepsilon_0}\left\{\frac{\omega_{0,i}^2 - \omega^2}{(\omega_{0,i}^2 - \omega^2)^2 + \gamma^2\omega^2}\right\}$$

$$\varepsilon''_{r,\infty} = \frac{N.q_i^2}{m_i\varepsilon_0}\left\{\frac{\gamma\omega}{(\omega_{0,i}^2 - \omega^2)^2 + \gamma^2\omega^2}\right\}$$

It is simple separation of variables. So, when you plot this now, what you will obtain is something like this. When you plot dielectric constant, so this is $\varepsilon_{r,\infty}$, as a function of frequency. We will see it goes, we just expand it around this point. So, this will be your **ω_0 (electronic)** . So, this is for electronic or it could be for **ω_0 (ionic)** depending upon the masses and charges.

And when you plot $\varepsilon''_{r,\infty}$, we will see at this point system shows there will be high loss. This is the reason why we see those resonances in the dielectric constant plot. So, we will dwell upon this a little bit more in the next lecture, but what we have established is the simple framework to calculate the dielectric constant, the complex dielectric constant on a microscopic scale for electronic and ionic polarization.