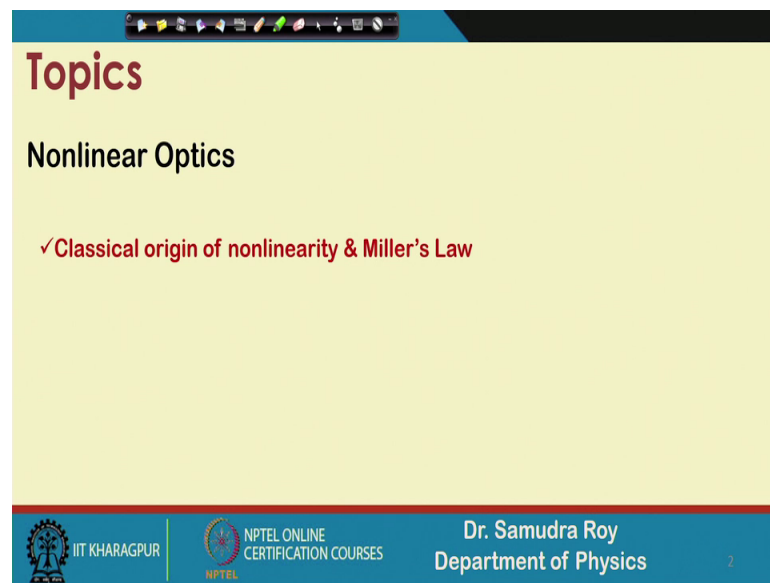


Introduction to Non-Linear Optics and its Applications
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Lecture - 12
Miller's Rule

So, welcome back student to the next class of Introduction to Non-linear Optics and its Application. So, this is lecture number 12. So, in lecture number 11 we have started a very important thing that the classical origin of nonlinearity. So, let us go back to whatever we have done in our previous class and make a recap of that.

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Topics

Nonlinear Optics

✓ Classical origin of nonlinearity & Miller's Law

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**Classical origin of nonlinearity
(Cont)**

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So, classical origin of nonlinearity is continued.

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Classical origin of nonlinearity

An-harmonic Oscillator model

$$U = \frac{1}{2}kX^2 + \frac{1}{3}amX^3$$

$$P = \epsilon_0\chi^{(1)}E + \epsilon_0\chi^{(2)}E^2 + \epsilon_0\chi^{(3)}E^3 + \dots$$

$$U = \frac{1}{2}kX^2 + \frac{1}{3}amX^3$$

The potential give rise to the force,

$$F = -m\omega_0^2X - maX^2$$

The equation of motion,

$$\ddot{X} + \Gamma\dot{X} + \omega_0^2X + aX^2 = qE/m$$

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So, that was our picture that if the electric field is very high, then electron will going to vibrate and this vibration will not be modelled as a harmonic oscillator, but some sort of an harmonic term should be there. And when we have some an harmonic term some kind of nonlinearity will be there in the system that the force will not be proportional to X anymore, it is now some XS got a components are also there.

So, as a result we have a different kind of equation of motion. And this equation of motion basically contain a non-linear term which is sitting here after that what we have done?

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The slide contains the following content:

- Graph:** A plot of potential energy versus displacement, showing a parabolic curve with a red dashed line representing the linear approximation and a solid red line representing the full potential with a non-linear term.
- Equation of motion:** $\ddot{X} + \Gamma\dot{X} + \omega_0^2 X + aX^2 = qE/m$
- Solution in power series expansion:** $X = \delta X^{(1)} + \delta^2 X^{(2)} + \dots$
- Order δ equation:** $\ddot{X}^{(1)} + \Gamma\dot{X}^{(1)} + \omega_0^2 X^{(1)} = qE/m$
- Order δ^2 equation:** $\ddot{X}^{(2)} + \Gamma\dot{X}^{(2)} + \omega_0^2 X^{(2)} = -a(X^{(1)})^2$

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After that let me go back to the same slide; that we have shown; that means, divide our solution as a series of as a power series where delta delta square are the corresponding coefficient or weightage of the solution. When we extract the weightage of delta and delta square we have 2 equation in our hand, one for X 1 and another for X 2. X 1 is nothing but our fundamental solution in absence of any kind of an harmonic term, and this is the same equation that we have been using for long. So, there is no an harmonic term present here.

But for X 2 which is interesting we have a source term and a is related to that which is related to the constant that gives rise to an harmonic term. So, next thing is to find out what is the solution of X 2.

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(Lecture 9)

$$\delta : \ddot{X}^{(1)} + \Gamma \dot{X}^{(1)} + \omega_0^2 X^{(1)} = qE/m$$

$E_1^{(\omega_1)}$

$E_2^{(\omega_2)}$

$$\ddot{x}_0 = \frac{q/m}{(\omega_0^2 - \omega^2 - i\gamma\omega)} E_0 = \frac{q/m}{D(\omega)} E_0$$

$$D(\omega) = (\omega_0^2 - \omega^2 - i\gamma\omega)$$

$$E = E^{(\omega_1)} + E^{(\omega_2)} = \frac{1}{2} [A_1 e^{-i\omega_1 t} + A_2 e^{-i\omega_2 t} + c.c.] \Rightarrow X^{(1)} = \frac{1}{2} [x_1^{(1)} e^{-i\omega_1 t} + x_2^{(1)} e^{-i\omega_2 t} + c.c.]$$

$$x_1^{(1)} = \frac{q/m}{D(\omega_1)} A_1$$

$$x_2^{(1)} = \frac{q/m}{D(\omega_2)} A_2$$

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In order to do that what we are doing that we launch the electric field and this electric field is now not a single electric field, but combination of electric field of different frequency ω_1 and ω_2 . When you do that; we find the solution x_1 and x_2 in this particular forms. These are the forms we are going to use please note that x_1 and x_2 form will be there. Why we are doing? Because my source term is now containing x_1 . So, we need to first find out what is my x_1 , x_1 was in my source term.

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$\delta^2: \ddot{X}^{(2)} + \Gamma \dot{X}^{(2)} + \omega_0^2 X^{(2)} = -a(X^{(1)})^2$
 $X^{(1)} = \frac{1}{2} [x_1^{(1)} e^{-i\omega_1 t} + x_2^{(1)} e^{-i\omega_2 t} + c.c.]$

(Driving Term)

$\ddot{X}^{(2)} + \Gamma \dot{X}^{(2)} + \omega_0^2 X^{(2)} = -a(X^{(1)})^2 = -\frac{a}{4} [x_1^{(1)} e^{-i\omega_1 t} + x_2^{(1)} e^{-i\omega_2 t} + c.c.]^2$

(Different frequency mixing)

(Frequency mixing)

$\omega_3 = \omega_1 + \omega_1 = 2\omega_1$
 $\omega_3 = \omega_2 + \omega_2 = 2\omega_2$
 $\omega_3 = \omega_1 + \omega_2$
 $\omega_3 = \omega_1 - \omega_2$
 $\omega_3 = \omega_1 - \omega_1 = \omega_2 - \omega_2 = 0$

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Well so that was our second equation and this equation suggests that we have some kind of source term sitting here depending on x_1 . And the next thing was what is the frequency there are many frequency component in the x_1 because the launching electric field was with 2 different frequencies and here we have the square term as a source because of this square term what happened? We have different frequency component.

What are the frequency component we have already listed here, and then eventually we find out from this term we find out what are the corresponding frequencies and what are the amplitude.

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$$\ddot{X}^{(2)} + \Gamma \dot{X}^{(2)} + \omega_0^2 X^{(2)} = -a(X^{(1)})^2 = -\frac{a}{4} \left[x_1^{(1)} e^{-i\omega_1 t} + x_2^{(1)} e^{-i\omega_2 t} + c.c \right]^2$$

↑
(Different frequency mixing)

$$(a + b + c + d)^2 = a^2 + b^2 + c^2 + d^2 + 2(ab + ac + ad + bc + bd + cd)$$

$$\left[x_1^{(1)} e^{-i\omega_1 t} + x_2^{(1)} e^{-i\omega_2 t} + x_1^{(1)*} e^{i\omega_1 t} + x_2^{(1)*} e^{i\omega_2 t} \right]_{(2\omega_1)}^2 = x_1^{(1)2} e^{-i2\omega_1 t}$$

$$\left[x_1^{(1)} e^{-i\omega_1 t} + x_2^{(1)} e^{-i\omega_2 t} + x_1^{(1)*} e^{i\omega_1 t} + x_2^{(1)*} e^{i\omega_2 t} \right]_{(2\omega_2)}^2 = x_2^{(1)2} e^{-i2\omega_2 t}$$

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We use simply a plus b plus c plus D whole square, and then we try to find out what are the different frequency components. If you look carefully you will find that omega 1 is a frequency 2 omega 1 is a frequency for which we have amplitude x 1 square 2 omega 2 is a frequency for which we have amplitude term x 2 whole square and so on.

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$$(a + b + c + d)^2 = a^2 + b^2 + c^2 + d^2 + 2(ab + ac + ad + bc + bd + cd)$$

$$\left[x_1^{(1)} e^{-i\omega_1 t} + x_2^{(1)} e^{-i\omega_2 t} + x_1^{(1)*} e^{i\omega_1 t} + x_2^{(1)*} e^{i\omega_2 t} \right]_{(0)}^2 = 2 \left(|x_1^{(1)}|^2 + |x_2^{(1)}|^2 \right)$$

$$\left[x_1^{(1)} e^{-i\omega_1 t} + x_2^{(1)} e^{-i\omega_2 t} + x_1^{(1)*} e^{i\omega_1 t} + x_2^{(1)*} e^{i\omega_2 t} \right]_{(\omega_1 + \omega_2)}^2 = 2x_1^{(1)} x_2^{(1)} e^{-i(\omega_1 + \omega_2)t}$$

$$\left[x_1^{(1)} e^{-i\omega_1 t} + x_2^{(1)} e^{-i\omega_2 t} + x_1^{(1)*} e^{i\omega_1 t} + x_2^{(1)*} e^{i\omega_2 t} \right]_{(\omega_1 - \omega_2)}^2 = 2x_1^{(1)} x_2^{(1)*} e^{-i(\omega_1 - \omega_2)t}$$

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So, in the next slide the same thing the 0 frequency omega 1 plus omega 2 frequency omega 1 minus omega 1 frequency. So, different frequency component i just extract that how the right hand side the amplitude will be there.

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The slide content is as follows:

$$\ddot{X}^{(2)} + \Gamma \dot{X}^{(2)} + \omega_0^2 X^{(2)} = -a(X^{(1)})^2 = -\frac{a}{4} \left[x_1^{(1)} e^{-i\omega_1 t} + x_2^{(1)} e^{-i\omega_2 t} + c.c. \right]^2$$

We chose this frequency $\omega_3 = \omega_1 - \omega_2$ (Different frequency mixing)

(Difference frequency)

$$\left[x_1^{(1)} e^{-i\omega_1 t} + x_2^{(1)} e^{-i\omega_2 t} + x_1^{(1)*} e^{i\omega_1 t} + x_2^{(1)*} e^{i\omega_2 t} \right]_{(\omega_1 - \omega_2)}^2 = 2x_1^{(1)} x_2^{(1)*} e^{-i(\omega_1 - \omega_2)t}$$

$$\ddot{X}^{(2)} + \Gamma \dot{X}^{(2)} + \omega_0^2 X^{(2)} = -\frac{a}{2} \left(x_1^{(1)} x_2^{(1)*} e^{-i\omega_3 t} + c.c. \right)$$

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And then finally, we have a frequency choose a frequency try to find out the value of x 2 for which, the frequency for which this is vibrating is omega 3 which is nothing but the difference between omega 1 and omega 2.

So, my equation here my governing equation here is this is my equation and the source term is reduced. So, there are many terms X 1 X 2 and complex conjugate square of that by I just extract only these things because I want to evaluate my x 2 x 2 value for the frequency omega 3 only. So, omega 3 is a frequency which is omega 1 plus omega 2. So, this is the thing we have done in the last class.

So, today we will proceed from here and try to find out what is the extra thing that we will going to get from this or what is the implication of this kind of solutions well.

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$$\ddot{X}^{(2)} + \Gamma \dot{X}^{(2)} + \omega_0^2 X^{(2)} = -\frac{a}{2} \left(x_1^{(1)} x_2^{(1)*} e^{-i\omega_3 t} + c.c. \right)$$

$$X^{(2)} = \frac{1}{2} x^{(2)} e^{-i\omega_3 t} + c.c. \quad (\text{Solution of the form})$$

$$(-\omega_3^2 + \omega_0^2 - i\Gamma\omega_3) x^{(2)} = -a x_1^{(1)} x_2^{(1)*}$$

Again in the box we write my differential equation for X_2 , which now depends on the amplitude of x_1 which is x_1 small x_1 1 small x_2 2 star of that an ω_3 some frequency component is also associated with that.

So, here we have one frequency in the source term and try to find out my x_2 . So, we will use the same old technique that my x_2 also should vibrate with the same frequency that of my source term. So, if we consider this is some sort of source which is vibrating at frequency ω_3 then; obviously, my x_2 will also going to vibrant at frequency ω_3 and we want the solution in this particular form, if I do then we will find what is the amplitude of x_2 we put this solution here simply. And then X double dot is nothing but everything same with just ω_1 ω_3 whole square term will be there ω_0 square will be as usual and then minus $i\Gamma\omega_3$ term that will appear because of this velocity dependent damping term and the right hand side we have only this term.

The half term will cancelled out because the half is here; also the half term is here. So, we will have a very compact form very compact form for the amplitude of X_2 which is vibrating as a frequency ω_3 .

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The slide displays the following equations and text:

$$\ddot{X}^{(2)} + \Gamma \dot{X}^{(2)} + \omega_0^2 X^{(2)} = -\frac{a}{2} \left(x_1^{(1)} x_2^{(1)*} e^{-i\omega_3 t} + c.c. \right)$$
$$\underline{X}^{(2)} = \frac{1}{2} x^{(2)} e^{-i\omega_3 t} + c.c. \quad (\text{Solution of the form})$$
$$(-\omega_3^2 + \omega_0^2 - i\Gamma\omega_3) x^{(2)} = -ax_1^{(1)} x_2^{(1)*} \checkmark$$

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So, x mind it X is a complete displacement having a frequency component this and x 2 is the corresponding amplitude. So now x 2 is following this equation and from here I can write x 2 in terms of D some parameter we have already used and this parameter D is read in terms of omega 1 omega 2 well.

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The slide displays the following equations and text:

$$(-\omega_3^2 + \omega_0^2 - i\Gamma\omega_3) x^{(2)} = -ax_1^{(1)} x_2^{(1)*} \checkmark$$
$$\underline{x}^{(2)} = \frac{-ax_1^{(1)} x_2^{(1)*}}{(-\omega_3^2 + \omega_0^2 - i\Gamma\omega_3)} = \frac{-ax_1^{(1)} x_2^{(1)*}}{D(\omega_3)}$$
$$D(\omega_3) = (-\omega_3^2 + \omega_0^2 - i\Gamma\omega_3) \checkmark$$

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So, here is a solution. So, we have this equation in my hand ω_3^2 this is the same equation that we have derived in the last slide. And from there if I want to find out my x_2 what we will do? This divided by this quantity that minus $a x_1 x_2^*$ divided by the inter stuff; that means, this one this one can write in the similar form that we have been writing that whenever $\omega^2 + \omega_0^2 - i\Gamma\omega$ these things are there we write this as a some function ω and everything can be written in terms of D . So, here D is this quantity so my x_2 is now written a very compact form which is this one.

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$$x^{(2)} = \frac{-ax_1^{(1)}x_2^{(1)*}}{(-\omega_3^2 + \omega_0^2 - i\Gamma\omega_3)} = \frac{-ax_1^{(1)}x_2^{(1)*}}{D(\omega_3)}$$

$$x_1^{(1)} = \frac{q/m}{D(\omega_1)} A_1 \quad x_2^{(1)} = \frac{q/m}{D(\omega_2)} A_2$$

$$x^{(2)}(\omega_3) = \frac{-aq^2 A_1 A_2^*}{m^2 D(\omega_1) D^*(\omega_2) D(\omega_3)}$$

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Well after that this is my x_2 and now the important thing we are going to do that, I like to this is a very important stuff this is a very important stuff because now what we will do I will replace my x_1 and x_2 in terms of these 2 solutions that we have been using. This solution as already we have in our previous calculation the first part of the calculation that what should be the what should be the value of x_1 and what should be the value of x_2 under any kind under any kind of an electric field launch electric field. So, launch electric field amplitude is A_1 and A_2 because there are 2 frequency components are there, for 2 different frequency components I have 2 displacements and these 2 displacement x_1 and x_2 can be written in this form this we have already done before.

So, the next thing is to replace the entire stuff here I just want to replace this x_1 and x_2 here if you do you will readily get this expressions. So, if I do so what should be the value?

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The slide contains the following mathematical expressions:

$$x^{(2)} = \frac{-ax_1^{(1)}x_2^{(1)*}}{(-\omega_3^2 + \omega_0^2 - i\Gamma\omega_3)} = \frac{-ax_1^{(1)}x_2^{(1)*}}{D(\omega_3)}$$

$$x_1^{(1)} = \frac{q/m}{D(\omega_1)}A_1 \quad x_2^{(1)} = \frac{q/m}{D(\omega_2)}A_2$$

$$x^{(2)}(\omega_3) = \frac{-aq^2A_1A_2^*}{m^2D(\omega_1)D^*(\omega_2)D(\omega_3)}$$

Handwritten annotations on the right side of the slide include:

$$x^{(2)} = -\frac{ax_1^{(1)}x_2^{(1)*}}{D(\omega_3)}$$

$$x^{(2)} = \frac{q^2/m^2A_1A_2^*}{D(\omega_3)D(\omega_2)}$$

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x_2 is minus of $a x_1 x_2^{(1)*}$ divided by D of ω_3 , x_1 is this quantity so I just replace x_1 . So, x_2 will be minus of $a x_1$ is nothing but q by m . So, there should be x_2 is also q by m . So, there will be $2 q$ by m . So, I should write q square divided by m square then whole divided by $1 A_1$ and $1 A_2$ will be there, but mind it x_2 is there. So, one will be A_1 and another will be A_2^* then whole divided by D of ω_3 as it is. And from here we have one D of ω_1 and another D of ω_2 , but since this is star so we should have a star here.

So, exactly the same equation we are getting here we are getting minus of $a q$ square divided by m square then multiplication of $A_1 A_2^*$ then divided by D of $\omega_1 D$ of ω_2 and D of ω_3 . D of ω_2 should have a star sign because I am replacing this x_2 to x_2^* which is here. Well so now everything x_2 is written in terms of $A_1 A_2$ which is the initial fields. So, that is interesting so; that means, I write the everything as a source term of x_1 and x_2 and x_2 again represented in terms of x_1 and x_2 is already there in terms of A_1 and A_2 which is the amplitude of the electric field

that we launch into the system.

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Slide content:

$$x^{(2)} = \frac{-ax_1^{(1)}x_2^{(1)*}}{(-\omega_3^2 + \omega_0^2 - i\Gamma\omega_3)} = \frac{-ax_1^{(1)}x_2^{(1)*}}{D(\omega_3)}$$

$$x_1^{(1)} = \frac{q/m}{D(\omega_1)}A_1 \quad x_2^{(1)} = \frac{q/m}{D(\omega_2)}A_2$$

$$x^{(2)}(\omega_3) = \frac{-aq^2(A_1A_2^*)}{m^2D(\omega_1)D^*(\omega_2)D(\omega_3)}$$

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And my x_2 is now written in terms of the amplitude of the launch electric field and also D omega terms that is here. Well so after having this compact form.

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Slide content:

$$x^{(2)}(\omega_3) = \frac{-aq^2A_1A_2^*}{m^2D(\omega_1)D^*(\omega_2)D(\omega_3)} \quad \checkmark$$

(2nd order nonlinear polarization)

$$P_{\omega_3}^{(2)} = -Nqx^{(2)} = \frac{\epsilon_0\chi^{(2)}}{\omega_3}A_1A_2^*$$

$$\omega_3 = (\omega_1 - \omega_2)$$

$$\chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \frac{Naq^3}{\epsilon_0m^2D(\omega_1)D(\omega_2)^*D(\omega_3)} \quad (\omega_3 = \omega_1 - \omega_2)$$

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We have something very interesting now because everything is they are in $D_1 D_{\omega_1}$ D_{ω_2} form, but $D_{\omega_2} D_{\omega_1}$ and D_{ω_3} which is again can be represented in terms of χ_2 . So, second order polarisation now this is x_2 this is my x_2 second order polarisation which is related to the displacement of x_2 this is the second order polarisation, first order polarisation if I write this is N_q simply x_1 .

But here the second order polarisation is related to the displacement of the x_2 is now can be represented as this quantity multiplied by the total field, multiplied by the total field, x_2 is related to this quantity. So, that is why; x_2 can be the total field can be now represented to this also one should write here this P_2 has a frequency component of ω_3 because P_2 can be vibrate in different frequencies, but from the very beginning we are considering a frequency ω_3 which is related to ω_1 and ω_2 like $\omega_1 - \omega_2$ this the difference frequency. So, for P_2 with a frequency ω_3 we have some term and this term is this and from where we can extract my χ_2 .

So, χ_2 can be now represented in terms of everything. So, I just replace here. So, these now equal to this. So, $A_1 A_2$ will be cancel out. So, ϵ_0 will go down and then there will be some kind of small calculation and if you do then you will come to from here to here you will come to this form.

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$$x^{(2)}(\omega_3) = \frac{-aq^2 A_1 A_2^*}{m^2 D(\omega_1) D^*(\omega_2) D(\omega_3)}$$

(2nd order nonlinear polarization)

$$P^{(2)} = -Nqx^{(2)} = \epsilon_0 \chi^{(2)} A_1 A_2^*$$

$\omega_3 = (\omega_1 - \omega_2)$ ↓

$$\chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \frac{Naq^3}{\epsilon_0 m^2 D(\omega_1) D(\omega_2)^* D(\omega_3)} \quad (\omega_3 = \omega_1 - \omega_2)$$

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So, chi 2 is a second order susceptibility please mind it that is arising because of this an harmonic term related to a. If I put a equal to 0 this term will vanish so there will not be any kind of chi 2.

Now, I try to find out what is the component of chi 2 frequency component which is omega 3. So, omega 3 is a frequency component of chi 2, how this frequency component is generated? You remember that omega 3 is generated as omega 1 minus omega 2 the different frequency. So, that is why here the notation is written and it is generated as omega 1 and another frequency component minus of omega 2 because of this minus sign.

And the right hand side I will have that this value is N a q cube then epsilon m square and this entire quantity, this entire quantity this quantity is already there. So, I have calculate my xi 2 in terms of D omega 1 D omega 2 and D omega 3 and the complex conjugate. So, 2 is having a complex conjugate so minus term is associative to omega 2. So, that is why for this D omega 2 we have the complex conjugate there is a easy way to understand.

We'll let us see what have? What we have in our next slide.

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$$\chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \frac{Naq^3}{\epsilon_0 m^2 D(\omega_1) D(\omega_2)^* D(\omega_3)}$$

$$\chi^{(1)}(\omega) = \frac{Nq^2/m\epsilon_0}{D(\omega)}$$

$$\chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \frac{ma\epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)*}(\omega_2) \chi^{(1)}(\omega_3)$$

So, this is a very, very interesting expression that we have actually. So, let us go back to our previous equation. So, this is the equation that we have explaining so far. So, chi 2 is a second order susceptibility which is at omega 3 related to 2 frequencies omega 1 and minus omega 2 and we are getting this.

But already we know that first order susceptibility is a frequency dependent term. And the first order susceptibility can be represented in this way this is this has already been done when we are learning the Lawrence model, the Lawrence model of susceptibility or the linear theory or the classical theory of susceptibility we have this expression in our hand, which is chi 1 is equal to N q square divided by m epsilon 0 by D omega.

Now, what we will do this quantity D omega is also sitting here right in terms of D omega 1 D omega 2 and D omega 3. So, what we will do we just replace this D in terms of chi 1 this is the very, very important consequence that if I do then what essentially we will get is very, very very, very interesting.

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And this interesting things that second order susceptibility having a frequency omega 3 and 2 different frequency omega 1 and omega 2 which is associated with that the combination of omega 1 and omega 2 basically gives rise to omega 3. It is it depends on the first order susceptibilities of those frequencies. If I replace omega. So, from here I can do very easily. So, my D omega 1 1 by D omega 1 will be how much chi 1 function of omega 1 divided by N of q square of m of epsilon 0.

Also m of D omega 2 star will be chi 1 star omega 2 divided by same quantity N q square divided by m epsilon 0. Now if I start replacing 1 by D omega 1 1 by D omega 2 and 1 by D omega 3, then we have N N N square and q to the power 6. So, N square N cube divided by q to the power 6, one N is sitting here N q cube is sitting here. So, this N will cancel out and we will have N square in the denominator and also we will have a q 3 term, because in the denominator we have q to the power 6 another term will come here and we have q to the power 6. So, this q to the power 6 will cancel out with this q cube term.

So, in the denominator we have q 3 term also m omega m epsilon 0 m epsilon 0 and m epsilon 0 term will there. So, we will have m square we will have in the numerator one term m cube epsilon 0 cube, but from here we can see in the numerator we have another

epsilon 0 m square term. So, eventually we have the epsilon square and m term in the numerator which is here and here a is already there. So, we will have so this is the term which we can calculate very easily.

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$$\chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \frac{Naq^3}{\epsilon_0 m^2 D(\omega_1) D(\omega_2)^* D(\omega_3)}$$

$$\chi^{(1)}(\omega) = \frac{Nq^2/m\epsilon_0}{D(\omega)}$$

$$\chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \frac{ma\epsilon_0^2}{N^2q^3} \chi^{(1)}(\omega_1) \chi^{(1)*}(\omega_2) \chi^{(1)}(\omega_3)$$

And the rest of the term is simply susceptibility the fundamental of the first order susceptibility at the frequency omega 1 omega 2 and omega 3 this is the very, very interesting expression.

So, this expression suggest that if you know the susceptibility of the frequency omega 1 omega 2 and omega 3, then you can able to say what should be the susceptibility of the second order susceptibility in omega 3. So, this is the relationship where the suspect second order susceptibility can be represented in terms of first order susceptibilities. Also one thing if you note that if you make the ratio of second order susceptibility.

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$$\chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \frac{Naq^3}{\epsilon_0 m^2 D(\omega_1) D(\omega_2)^* D(\omega_3)}$$

$$\chi^{(1)}(\omega) = \frac{Nq^2/m\epsilon_0}{D(\omega)}$$

$$\chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \frac{ma\epsilon_0^2}{N^2q^3} \chi^{(1)}(\omega_1) \chi^{(1)*}(\omega_2) \chi^{(1)}(\omega_3)$$

$\frac{\chi^{(2)}(\omega_3; \omega_1, -\omega_2)}{\chi^{(1)}(\omega_1) \chi^{(1)*}(\omega_2) \chi^{(1)}(\omega_3)} = \frac{ma\epsilon_0^2}{N^2q^3}$

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For example, I am making a ratio omega 3 which is coming as actually frequency omega 1 and another frequency minus of omega 2 divided by the first order susceptibilities. Susceptibilities of first order at different frequencies with a star term here at omega 3 this ratio is m a epsilon 0 square then N square q cube.

So, this quantity m a epsilon 0 square m square q cube is a constant. So, if you make a ratio of the second order susceptibility at q 3 and all this first order susceptibility related to all this frequencies this ratio is always become a constant term. This is a very important thing and we have a name for that.

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Miller's Rule

An empirical rule which states that the 2nd order susceptibility is proportional to the product of the 1st order susceptibilities at three frequencies which 2nd order susceptibility is depend upon.

$\omega_3 = \omega_1 + \omega_2$

For Sum frequency generation ($\omega_3 = \omega_1 + \omega_2$),

$$\chi^{(2)}(\omega_3; \omega_1, \omega_2) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)}(\omega_2) \chi^{(1)}(\omega_3)$$

For Difference frequency generation ($\omega_3 = \omega_1 - \omega_2$),

$$\chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)*}(\omega_2) \chi^{(1)}(\omega_3)$$

For Second harmonic generation ($\omega_3 = \omega_1 + \omega_1 = 2\omega_1$),

$$\chi^{(2)}(2\omega_1; \omega_1, \omega_1) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)}(\omega_1) \chi^{(1)}(2\omega_1)$$

For optical rectification ($\omega_3 = \omega_1 - \omega_1 = 0$),

$$\chi^{(2)}(0; \omega_1, -\omega_1) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)*}(\omega_1) \chi^{(1)}(0)$$

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Which is called the Millers rule this rule is called the Millers rule. So, what Millers rule suggest the Millers rule is an empirical rule will state that the second order susceptibility is proportional to the product of the first order susceptibilities at 3 frequencies which second order susceptibility is depend upon.

So, if I go back so this is the second order susceptibility, second order susceptibility I try to find out what is the value of the second order susceptibility at particular frequency. So, this particular frequency is omega 3 omega 3 frequency can be figure out by omega 1 and omega 2 in our calculation.

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$$\chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \frac{Naq^3}{\epsilon_0 m^2 D(\omega_1) D(\omega_2) D(\omega_3)}$$

$$\chi^{(1)}(\omega) = \frac{Nq^2/m\epsilon_0}{D(\omega)}$$

$$\chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \frac{ma\epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)*}(\omega_2) \chi^{(1)}(\omega_3)$$

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We consider omega 3 is omega 1 minus omega 2 that was our consideration. When we take omega 3 is equal to omega 1 minus omega 2 3 different frequency components are there that is associated with this.

So now what happen that this is now proportional to the first order susceptibility of this frequency, first order susceptibility of this frequency and the first order susceptibility of this frequency. So, this is a very, very interesting outcome that we have and this is called the Millers rule. So, let us go back to the Millers rule and try to find out more thing. So, Millers rule as I mentioned it is suggest that this is a empirical rule which states that the second order susceptibility is proportional.

So now we will try to find out what should be the form of the Millers rule, what should be the form of the Miller Millers rule when we have different frequency components. So now, we have a frequency component omega 1 minus omega 2, but as I mentioned there may be some other frequency dependent component frequency components also.

For example, if my susceptibility is related to omega 1 and omega 2; that means, omega 3 is represented by omega 1 plus omega 2 sum of these 2 frequencies, I can extend this also this frequency also. Then according to Millers rule this constant coefficient will be

same which is called the Millers coefficient and then susceptibility at omega 1 susceptibility at omega 2 and susceptibility at omega 3 will be there. Mind it since I am working with the plus of omega 2 there is no star sign here, which was there in the difference frequency generation.

In the different frequency generation Millers rule suggest that susceptibility second order susceptibility at some frequency omega 2, which is omega 3 which is related to omega 3 as omega 1 minus omega 2 this things.

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Miller's Rule

An empirical rule which states that the 2nd order susceptibility is proportional to the product of the 1st order susceptibilities at three frequencies which 2nd order susceptibility is depend upon.

$\omega_3 = \omega_1 - \omega_2$

For Sum frequency generation ($\omega_3 = \omega_1 + \omega_2$),

$$\chi^{(2)}(\omega_3; \omega_1, \omega_2) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)}(\omega_2) \chi^{(1)}(\omega_3)$$

For Difference frequency generation ($\omega_3 = \omega_1 - \omega_2$),

$$\chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)*}(\omega_2) \chi^{(1)}(\omega_3)$$

For Second harmonic generation ($\omega_3 = \omega_1 + \omega_1 = 2\omega_1$),

$$\chi^{(2)}(2\omega_1; \omega_1, \omega_1) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)}(\omega_1) \chi^{(1)}(2\omega_1)$$

For optical rectification ($\omega_3 = \omega_1 - \omega_1 = 0$),

$$\chi^{(2)}(0; \omega_1, -\omega_1) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)*}(\omega_1) \chi^{(1)}(0)$$

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This will be proportional to the first order susceptibility at omega 1, first order susceptibility at omega 2, but with a negative sign because omega 2 has a negative sign. And also the first order susceptibility at omega 3 which is the difference between these 2 frequency.

If I multiply these 3 things and then multiply my constant which is called the Millers constant, then I am going to have the second order susceptibility at that particular frequency. So, obvious with the frequency is important thing, if an if I try to find out the second order susceptibility I need to find out at which frequency I am talking about there are a few frequencies for which, we will find the second order susceptibility. And here

some frequency difference frequency and second order frequencies are there second harmonic frequencies are also there. So, let us see how the second harmonic frequencies are there.

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Miller's Rule
An empirical rule which states that the 2nd order susceptibility is proportional to the product of the 1st order susceptibilities at three frequencies which 2nd order susceptibility is depend upon.

Handwritten note: $\omega_3 = \omega_1 + \omega_1$

For *Sum frequency generation* ($\omega_3 = \omega_1 + \omega_2$),

$$\chi^{(2)}(\omega_3; \omega_1, \omega_2) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)}(\omega_2) \chi^{(1)}(\omega_3)$$

For *Difference frequency generation* ($\omega_3 = \omega_1 - \omega_2$),

$$\chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)*}(\omega_2) \chi^{(1)}(\omega_3)$$

For *Second harmonic generation* ($\omega_3 = \omega_1 + \omega_1 = 2\omega_1$),

$$\chi^{(2)}(2\omega_1; \omega_1, \omega_1) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)}(\omega_1) \chi^{(1)}(2\omega_1)$$

For *optical rectification* ($\omega_3 = \omega_1 - \omega_1 = 0$),

$$\chi^{(2)}(0; \omega_1, -\omega_1) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)*}(\omega_1) \chi^{(1)}(0)$$

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Second harmonic frequency means omega 3 is ready to omega 1 and omega 1. So, there is a possibility that omega 3 will generate to 2 omega 1 omega 1 plus omega 1. So, that is the case then we have the second order susceptibility at 2 omega 1. So, in that case we will write x 1 omega 1 susceptibility chi 1 at omega 1 chi 1 at omega 1 and chi 1 at 2 omega 1.

One can write also chi 1 at omega 1 square of that this is the same thing. Also for omega 2 frequency also we have the same thing.

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Miller's Rule

An empirical rule which states that the 2nd order susceptibility is proportional to the product of the 1st order susceptibilities at three frequencies which 2nd order susceptibility is depend upon.

For Sum frequency generation ($\omega_3 = \omega_1 + \omega_2$),

$$\chi^{(2)}(\omega_3; \omega_1, \omega_2) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)}(\omega_2) \chi^{(1)}(\omega_3)$$

For Difference frequency generation ($\omega_3 = \omega_1 - \omega_2$),

$$\chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)*}(\omega_2) \chi^{(1)}(\omega_3)$$

For Second harmonic generation ($\omega_3 = \omega_1 + \omega_1 = 2\omega_1$),

$$\chi^{(2)}(2\omega_1; \omega_1, \omega_1) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)}(\omega_1) \chi^{(1)}(2\omega_1)$$

For optical rectification ($\omega_3 = \omega_1 - \omega_1 = 0$),

$$\chi^{(2)}(0; \omega_1, -\omega_1) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)*}(\omega_1) \chi^{(1)}(0)$$

Handwritten notes in blue ink:

$\omega_3 = \omega_1 + \omega_2$

$\chi^{(2)}(2\omega_1; \omega_1, \omega_1) = \left(\frac{m a \epsilon_0^2}{N^2 q^3} \right) \chi^{(1)}(\omega_1) \chi^{(1)}(\omega_1) \chi^{(1)}(2\omega_1)$

So; that means, omega 3 can also be written as omega 2 plus omega 2. In this case this things will simply be replaced like susceptibility at 2 omega 2, which is related to 2 frequencies omega 2 and omega 2. And in the right hand side I should write it is simply m a epsilon 0 square divided by N square q cube, which is the Millers coefficient or Millers constant. And then I write omega of ah chi of 1 at omega 2 square of that because there will be 2 term like this and then another term chi of 1 at 2 omega 2 this is the thing. In a similar way we have optical rectification term.

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Miller's Rule

An empirical rule which states that the 2nd order susceptibility is proportional to the product of the 1st order susceptibilities at three frequencies which 2nd order susceptibility is depend upon.

For Sum frequency generation ($\omega_3 = \omega_1 + \omega_2$),

$$\chi^{(2)}(\omega_3; \omega_1, \omega_2) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)}(\omega_2) \chi^{(1)}(\omega_3)$$

For Difference frequency generation ($\omega_3 = \omega_1 - \omega_2$),

$$\chi^{(2)}(\omega_3; \omega_1, -\omega_2) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)*}(\omega_2) \chi^{(1)}(\omega_3)$$

For Second harmonic generation ($\omega_3 = \omega_1 + \omega_1 = 2\omega_1$),

$$\chi^{(2)}(2\omega_1; \omega_1, \omega_1) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)}(\omega_1) \chi^{(1)}(2\omega_1)$$

For optical rectification ($\omega_3 = \omega_1 - \omega_1 = 0$),

$$\chi^{(2)}(0; \omega_1, -\omega_1) = \frac{m a \epsilon_0^2}{N^2 q^3} \chi^{(1)}(\omega_1) \chi^{(1)*}(\omega_1) \chi^{(1)}(0)$$

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In optical rectification term there should be any frequency components. So, $\omega_1 - \omega_2$ or $\omega_2 - \omega_1$ this term will be there. So, $\omega_1 - \omega_1$ give rise to ω_0 . So, I can also have my susceptibility which should have any kind of frequency component, and that can one can figure out which susceptibility at ω_1 susceptibility star at ω_1 and susceptibility at 0 frequencies.

So, you can see that Millers rule suggest that if you know the susceptibility first order susceptibility at some frequencies, then by combining this frequencies we can generate the susceptibility second order susceptibility. And you need just need to multiply some constant multiplier. So, with this note I like to conclude.

So, today we will learn a very important thing that how classically the non linearities generated the non linearities generated because of the fact that, if I launch electric field which is very high then the electron that will going to vibrate based on the Lawrence model the electron that will going to vibrate, will have some kind of an harmonicity because of this an harmonicity what happen? We have some additional term which is depend on x^3 in the potential.

So, this x^3 potential term basically gives to some kind of force which is square of that. So, force become non-linear and when we solve the differential equation for that particular force we will find the solution and this solution is a second order solutions. So,

second order solutions gives us the fact that we will there is a generating term related to the displacement first order displacement. And if you do the calculation carry on the calculation the second order displacement basically leads to the second order susceptibility term from the polarisation concept. And eventually we will get the relationship between the second order polarisation or the second order susceptibility to the first order susceptibility to the Millers rule.

So, with this note let me conclude here. So, see you in the next class so in the next class again we will going to discuss about the non-linearity. So, we will start learning how the second harmonic will going to generate and the for the second harmonic generation we need something for the second harmonic generation how 2 different wavelengths are launch. And for 2 different wavelengths if 2 different wavelengths are same that is ω_1 and ω_2 we can generate $2\omega_1$. And how this $2\omega_1$ generation can be can be can be appreciated by some mathematical simple mathematical treatment we will learn that. So, with this note let me conclude here.

Thank you very much for attention. So, see you in the next class.