Electrochemical Impedance Spectroscopy
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Lecture - 41
NLEIS. Introduction and Mathematical Background

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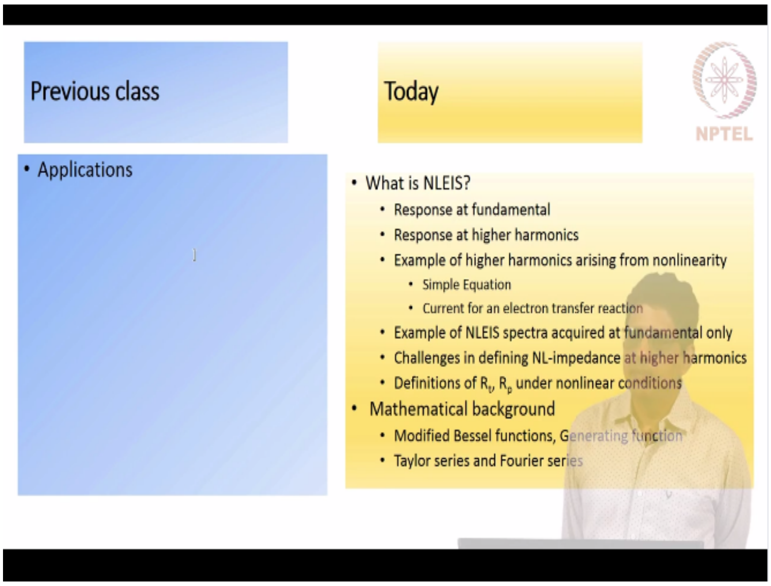

So earlier we have seen some applications okay. Now I want to introduce a technique called nonlinear EIS okay. So we normally measure the response at the fundamental that is we apply a potential sinusoidal wave at some frequency and measure the current at that frequency okay. It is possible to conduct this measurement or perform these measurements at higher harmonics; it is also possible to increase the amplitude.

So far we have assumed that the perturbation that is if I send a sine wave then the amplitude is small maybe 5 millivolts, maybe 10 millivolts but we can linearize the equation and analyze that is what we have assumed. What if we send a larger amplitude or what if we apply a larger amplitude perturbation? So I want to go through the description of nonlinear EIS, what is meant by nonlinear EIS and introduction to the terminology and what happens if you acquire the spectrum at large amplitude for a simple reaction okay.

We will go through the description or the calculations at a later stage. Right now, I just want to show you and I also want to describe what would be the corresponding values for a charge transfer resistance and polarization resistance if we apply large amplitude perturbations. It needs a certain level of mathematics to understand nonlinear EIS. So I will spend some time on giving you the background mathematical background necessary for this.

So it uses what is called modified Bessel functions, so we will spend some time on that and we will also look at Taylor series expansion for a small change when the change is described by a sine wave okay and we will compare with Fourier series expansion. So you should be familiar with Taylor series, Fourier series and some special functions okay. This helps you understand nonlinear EIS and helps you understand and use it well.

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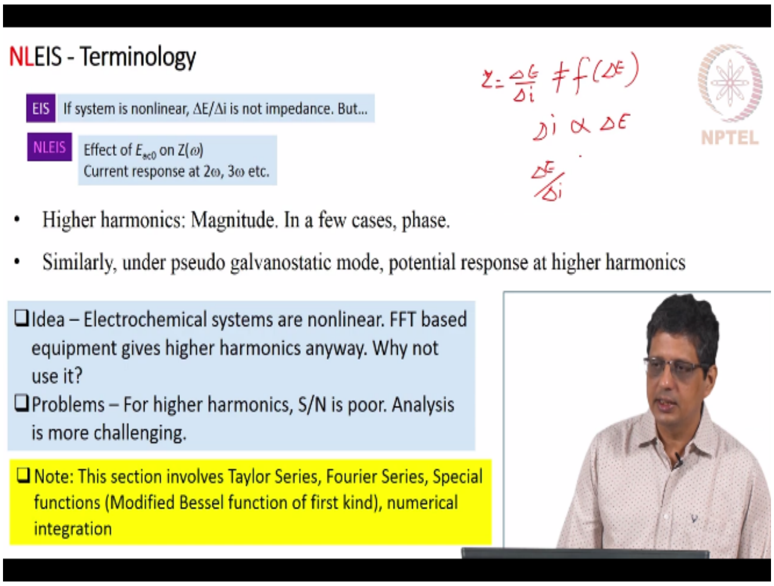

So first impedance is basically the ratio, vector ratio of potential to current, we apply a small amplitude perturbation del E and we monitor the current or change in current del i and that is differential impedance, we normally call it as impedance okay and that ratio of del E/del i that is actually not a function of del E that is the assumption. Del i is proportional to del E that is another way of saying it okay and this is valid if the system is linear.

If the system response cannot be linearized, then strictly speaking this is not a correct definition of impedance. It is just the transfer function; it cannot be called as impedance but then lot of people use the terminology nonlinear EIS to indicate the ratio of del E/del i. Then, it is a function of del E. Suppose I applied 5 millivolts perturbation; 5 millivolts sine wave and I get a impedance spectrum.

If I apply 10 millivolts and if I get pretty much the same impedance spectrum, then I can say that 5 millivolts or 10 millivolts, this system gives a linear response okay. If I go to 50 millivolts, if I go to 100 millivolts perturbation, then the spectrum that I get may be different in which case I should say nonlinear effects are incorporated okay. You can also measure the current response at higher harmonics.

If I apply at 5 Hertz frequency, I will get signal at 5 Hertz, I might also get signal at 10 Hertz, 15 Hertz, 20 Hertz okay. When the system response is in the linear regime, those signals at the higher harmonics 10 Hertz, 15 Hertz, etc they will be negligible. When the system response is in the nonlinear domain, then we will expect significant signal at those frequencies.

For example, if I apply 100 millivolts sine wave, a sine wave of 100 millivolt amplitude with 5 Hertz as the frequency, I will get a response at 5 Hertz but I will also get some response at 10, 15, 20 Hertz okay. Most of the times when researchers acquire data at higher harmonics, most of the times they would present magnitude data, only in few cases they present the phase data.

Basically, it becomes difficult to get good quality data especially the phase. So, so far I have described examples with pseudo-potentiostatic mode that means we apply a dc bias or we may not apply any dc bias, zero bias. We control the potential and monitor the current and then you can measure at higher harmonics, you can measure at the fundamental and then use this data for analysis.

You can also use current as applied input that is you can use under pseudo galvanostatic mode where you can use a zero or nonzero dc bias for the current and then apply a sinusoidal wave in the current and measure the potential okay and then you can take the ratio with the ratio of the potential to current and that will also give us the nonlinear EIS data. So the idea behind applying NLEIS is this.

Electrochemical systems, we know by and large you are going to get nonlinear response, the systems are nonlinear and a lot of times we used FFT based equipment or instrument to obtain and analyze the data. So FFT based instrument would anyway give us the response at fundamental as well as at higher harmonics. So why not use them okay, so that is idea. We do not see as many publications with NLEIS as you see with EIS.

EIS is used extensively whereas NLEIS is used only in very few cases. The main reason is the analysis is challenging okay, even at fundamental if we apply a large amplitude perturbation, we will get good quality data that means signal-to-noise ratio will be good. So it is not that difficult to get NLEIS data especially at fundamental but calculating the NLEIS response for a given model, given reaction kinetics that is more difficult.

So experimentally acquiring data is not that difficult. If you want to acquire at higher harmonics, you need some effort but acquiring at fundamental it is easy but analyzing the data is harder and that is the reason it is not seen that frequently and if it comes to higher harmonics, the signal-to-noise ratio is poor, usually it is poor. So you do not get good quality data that easily and then analysis is anyway difficult so it is less frequent.

So as I mentioned earlier, NLEIS involves a good understanding of Taylor series. If you have a good understanding of Taylor series, Fourier series and special functions and numerical integration methods using a software, this will actually help us get the maximum out of NLEIS technique.

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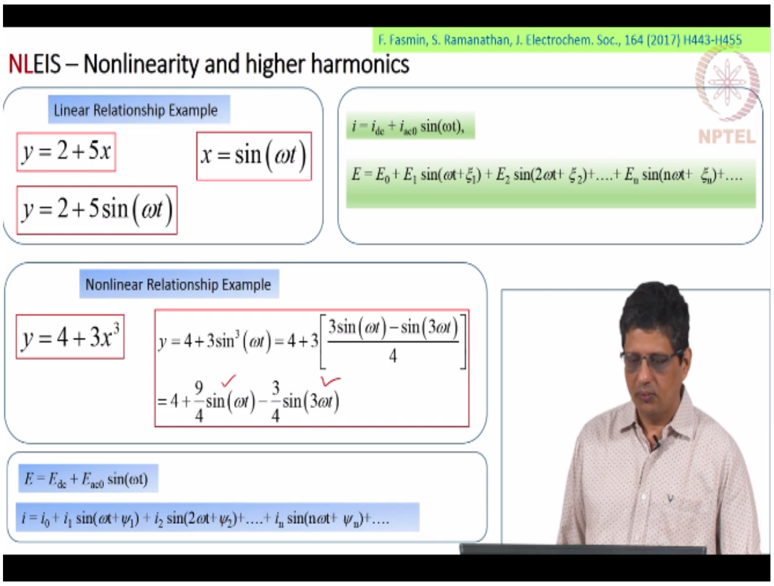

So I will start with some example, simple examples okay. You have one relationship, y=2+5x and if I draw y versus x, it is going to be a straight line okay and another expression is y=4+3x cube, I just made up this expression and this is going to give your curve. If you plot y versus x in a reasonable range, you would see a curve, 0 to 10 for example of x value going from 0 to 10, y value go from whatever numbers and you would not see a straight line, you would see a curve.

And let us say x is replaced with sine omega t okay, y is going to give you a sine omega t that means if I apply a potential which is a sine wave, the current response here given by y is also a sine wave. What happens when I look at an expression which has a nonlinear relationship okay? X cube, now we replace x cube with sine cube and sine cube can be written using the trigonometric identity as 3 sin omega t-sin 3 omega t and all of these divided by 4.

You can substitute and rearrange, you would find that you have applied a sine wave, you would get a response at omega, you would also get a response at 3 omega, that means we apply a frequency of 5 Hertz, we will get response at 5 Hertz, will also get a response at 15 Hertz for this particular expression. If you have another nonlinear expression, you might get at 10 Hertz; you might get at 20 Hertz okay.

You might get at 10, 20, 15 all those frequencies okay. So it is just to indicate that the relationship between potential and current are nonlinear then you should expect higher harmonics. The strength of the signal, the amplitude of the higher harmonics may be small but if the system is nonlinear, you should expect a high harmonics.

So what we have done so far is assume that we are applying only a small amplitude perturbation so the strength of the higher harmonics are negligible and the ratio of response at the fundamental okay, the del i to del E or del E/del i, either one of them is independent of del E. This is the assumption we have made so far.

So if I apply a dc potential and on top of it superimpose an ac potential and that is given by E here, then in general I should expect a response current response with the dc plus a response at fundamental with possibly a phase and a response at second harmonic with some phase, third harmonic with some phase. In this particular example, you have seen zero phase but in general you can expect some phase for each of these.

So we would call i1 as response at fundamental, i2 as response at second harmonic, i2 along with psi 2 okay. In the same way, you can also use this in galvanostatic mode or pseudo galvanostatic mode where the applied current is given by idc+iac0 sin omega t and the measured potential can be written in Fourier series with the dc offset response at fundamental, response at second harmonic, etc. For each of this, you have an amplitude under phase okay.

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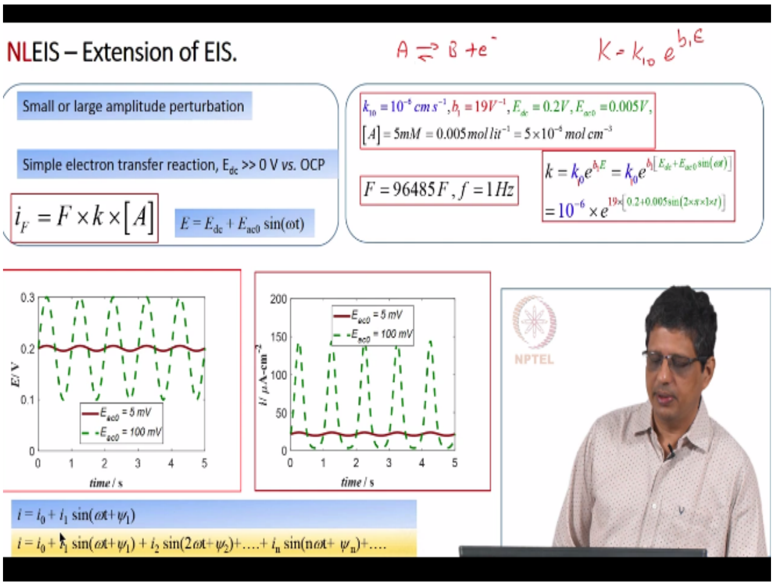

Now if I take a simple electron transfer reaction okay, electron transfer reaction is something like A going to B with an electron okay. If I apply a large positive potential with respect to open circuit potential, I can ignore the reverse reaction and I can assume that pretty much only the forward reaction happens. Under those conditions, I can write the current as faradaic current as Faraday constant, rate constant k and the concentration of the species A.

We will assume that mass transfer is not a limitation, it is very rapid. So we apply a dc potential and on top of it we have applied an ac potential. I have made up some numbers here for certain rate constant value 10 power -6 centimeters per second and for this we have used this expression k10 e power b1 E. So the exponent b1 is 19 Volt inverse. The dc potential for example in this case, I had taken as 0.2 Volts, 5 millimolar is the concentration of A and we apply a 5 millivolts ac potential.

If I apply 5 millivolt ac potential, I can calculate the value of k for these k0 or k10 and b1 and this dc potential and I have taken the example at 1 Hertz. The potential versus time would look like this, a 5 milli Volts ac would look like this and the current in micro amps per centimeter square would also appear as a sinusoidal current and there is no phase difference, you would get an impedance.

For the same reaction, I can write this as i0+i1 sin omega t+phi 1 or +psi 1 and psi value is 0, i1 I can calculate the value, I can take the Fourier transform and calculate the value and pretty much I can say this is sinusoidal wave for the current. What happens if I apply 100 millivolts AC, 100 millivolts amplitude of AC? The potential looks sinusoidal and with the same frequency of course.

The current response is definitely not sinusoidal, it is not a simple sine, it is nonlinear response and if you do Fourier transform, you would find that this can be written as sum of many sines with a dc offset and this should be written as i0+i1 sin omega t+psi 1 i2 sin 2 omega t+psi 2, etc. That means the response contains second, third, fourth harmonics. So this is definitely a nonlinear response.

This is just to show you that you would expect a nonlinear response when you use large amplitude perturbation and we have taken a very simple reaction and we have assumed only the forward reaction is significant, reverse reaction is neglected because we are at a dc bias where forward reaction would happen and reverse reaction is not going to happen.

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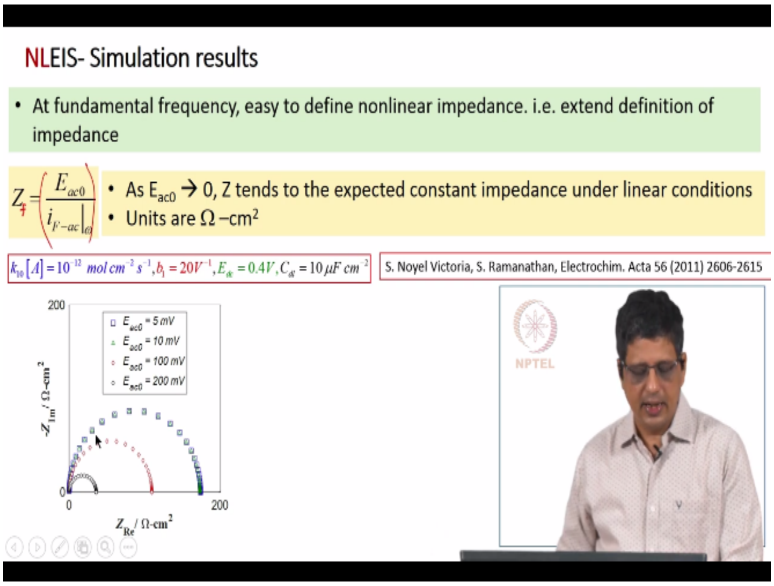

Definition of impedance is del E/del i and when we look at the response at fundamental then it is easy to define nonlinear impedance. I would say nonlinear impedances Eac0 by iac at omega. So this is faradaic impedance is at F when Eac0 is small and the response contains only response at fundamental and not at higher harmonics, then this is actually the correct definition of impedance.

When Eac0 is large and the response current response contains response at fundamental and at higher harmonics; we can still take the response at fundamental and take this ratio, this in that case Z F, NL that is going to be a function of Eac0 okay. These units are going to be ohm centimeter square, the current is current density actually, so it is going to be ohm centimeter square for this impedance.

And when Eac0 becomes smaller and smaller, if I start with 100 millivolts perturbation, I decrease it to 50 millivolts, the impedance response will be little different; decrease it to 20 millivolts, it is going to be different; from 20 millivolts to 10 millivolts, it would not be that much different for a given set of parameters okay. When I go to 1 millivolt, it is pretty much going to be independent of the applied perturbation.

So as Eac0 tends to small values, tend towards 0, Z tends towards the expected value, expected meaning when we expect it to be under linear regime. So it is going to the constant impedance value for a given frequency. Now it is easy to extend this and say okay if I go to higher amplitude, I will still get a number except that it is going to depend on this Eac0 and that number I would call as nonlinear impedance.

We will measure for the entire system, so we will call that as nonlinear impedance of the system. For example, for a simple electron transfer reaction with a slightly different set of values for these parameters with another dc bias and with an assumed double layer capacitance, we have simulated or calculated the nonlinear impedance. Later, we will learn how to calculate this, right now just take it for granted that this is correct.

So for this system if we apply 5 millivolt AC, we would get a spectrum that is given by the blue color squares and blue color squares as well as green triangles are more or less merged together or more or less identical. So if I apply 5 millivolt or 10 millivolt, I get the same impedance spectrum. That means this system; the response is still in the linear regime when we are at 5 or 10 millivolts.

If I go to 100 millivolts, the results that you would get are given by the red color diamonds, so it is going like this. It is definitely different from what we see at 5 millivolts and 10 millivolts. So we would say this is a nonlinear response. We are still measuring at the fundamental that means we apply 5 Hertz; we look at the response at 5 Hertz. We ignore the response at other frequencies. We ignore the response at dc.

We applied 20 Hertz; we look at the result at the same frequency. We apply 1 millihertz, we look at the result of the same frequency and then of course when you go to 200 millivolts, it is quite different, it is given by the dark circles or black circles and you can definitely say up to 10 millivolts, this is in the linear regime, beyond that it is in the nonlinear regime okay.

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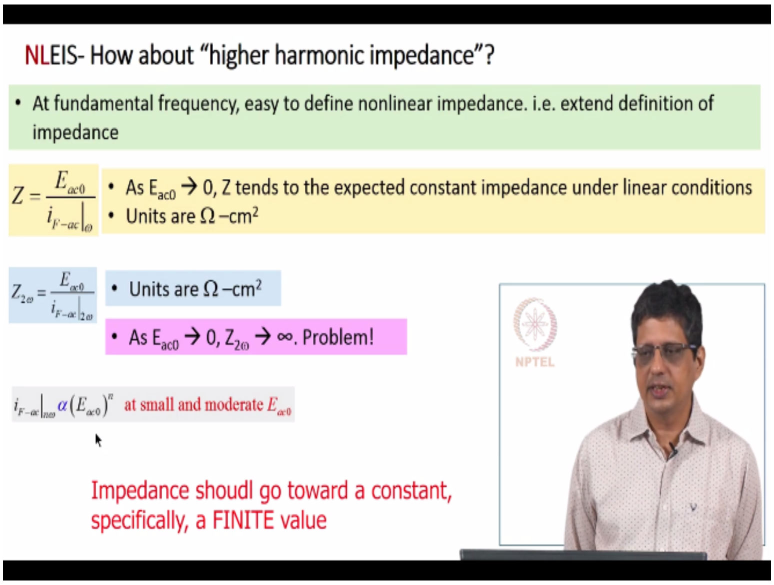

Okay so we are able to define or we are able to come up with a definition of nonlinear impedance when we look at the response at fundamental. When we go to higher harmonics, there is some problem okay. So I will describe this. Let us say we measure the current at fundamental, we also measure the current at second harmonic and we call that as iF ac 2 omega, we do not measure the faradaic current; we measure the total current okay.

But right now we are looking at the definition; we might be able to calculate the faradaic current for a given model or a given mechanism. So we can still say alright I will take the ratio of Eac0 by iF ac or i total ac at second harmonics 2 omega. Take the ratio, the good thing is that also has the units of ohm centimeter square, so we can call it as impedance. When you look at impedance, you would expect it to have units of ohm or ohm centimeter square.

Ohm for a normal electrical system, for electrode it is usually described as ohm centimeter square or usually given in ohm centimeter square but the problem is this, when we go to smaller and smaller value of Eac0 okay, this definition of Z tells us that the Z will go towards infinity and that is not the same way you would see for a response at fundamental okay. What you want to see is ideally a case that the impedance units are ohm or ohm centimeter square.

And when you go to lower and lower amplitude or smaller and smaller amplitude, that impedance should go towards a constant for a given frequency okay. However, you go to higher harmonics, the higher harmonic currents iac it need not be iF, i total or iF ac at n omega, second, third, fourth can be 1, can be 2, 3, 4, etc n is an integer that is proportional to Eac power n for small and moderate Eac0.

When you go to very low Eac0, it would not be proportional to this; we will see that again later okay. At moderate values where we expect to see reasonable signal with a good signal-to-noise ratio, this is the case. If you go to very large Eac0, you would get good signal-to-noise ratio but then it is going to be proportional to power which is more than m. So we would say it is definitely going to be at least as high as Eac0 power n.

And then if I take this ratio I would find that it is Eac0/Eac0 square multiplied by some constant. So when we go to small value of Eac0, this is going to be tending towards infinity. If you go to third harmonic, fourth harmonic, it will tend towards infinity at a more rapid rate.

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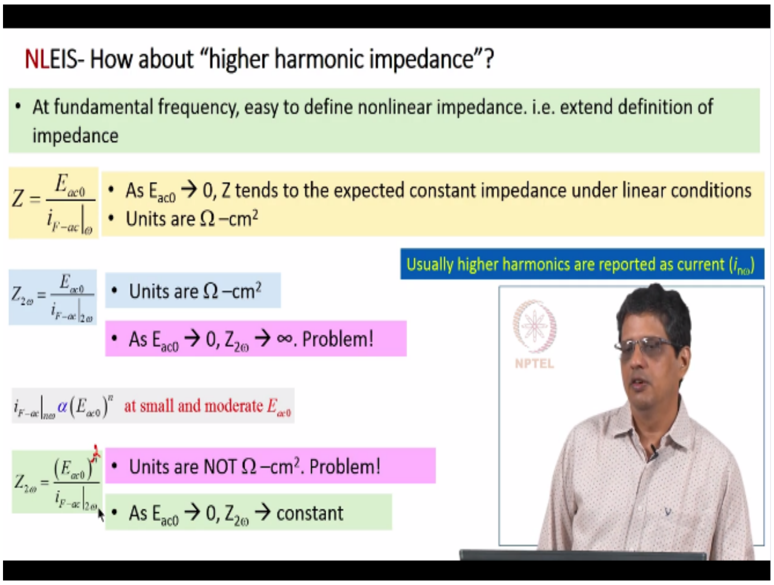

I can come up with another definition that is to say impedance at higher harmonics are nth harmonic can be Eac0 power n and then divide by iac at n omega. In this case, I should write it as Eac0 power 2 or Eac0 power n/n omega and that is going to be Zn omega. Problem is units are not ohm centimeter square. The units will depend on what harmonic we are using okay, so neither of them is very pleasant.

So generally in literature, although people say nonlinear EIS, they would frequently report only the current response at higher harmonics. At fundamental, you can give the current response; you can also give that as impedance. So usually higher harmonics are reported as current when we use pseudo-potentiostatic mode.

Of course, when we use pseudo galvanostatic mode, we will measure the potential and then subject it to Fourier transform, get the coefficients and then report the magnitude and phase, mostly magnitude, sometimes magnitude and phase at higher harmonics for the potential. We do not report the ratio of E to i or ratio of E power n to i for this.

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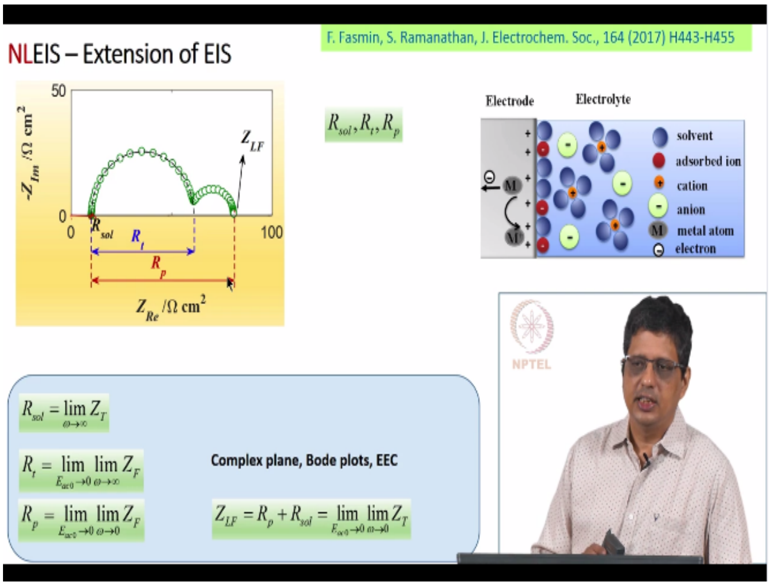

So to refresh your memory, we have seen few terms, solution resistance, charge transfer resistance and polarization resistance. If we take an electrode system, this is one example. If we have a system with one adsorbed intermediate, a clean system with rapid mass transfer, you can get a data like this. This is in complex plane plot, at high frequencies; the total impedance of the system gives us what is called as R solution.

So we would write solution resistance as total resistance Z total as omega tends to infinity. Charge transfer resistance is in case we are able to model this data and we are able to isolate and find the impedance of the faradaic reaction. What would be the impedance of the faradaic reaction when frequency tends to infinity, not for the total system, only for the faradaic component okay?

At the infinite frequency limit and at the linear regime, that means Eac0 should be very small, then we call it as charge transfer resistance. Previously, when we defined it, we would have just written as Rt is limit omega tending to infinity ZF. Reason is we were always assuming that we are operating in the linear regime; we did not have to explicitly say that each time but now we are dealing with cases where the Eac0 or the perturbation need not be small and sometimes it is deliberately made as a large value.

Therefore, we want to say or differentiate between cases where it is in the linear regime and where it is in the nonlinear region. So charge transfer resistance is at small amplitude perturbation, high frequency limit of faradaic component. Polarization resistance is small amplitude perturbation, low frequency limit omega tending to zero the faradaic component.

What we measure is of course the total resistance or total impedance, so if I take the low frequency limit, this is ZLF, this is the sum of solution resistance and polarization resistance okay. Now this is the definition of polarization resistance, charge transit resistance, etc with the assumption that the perturbation is small.

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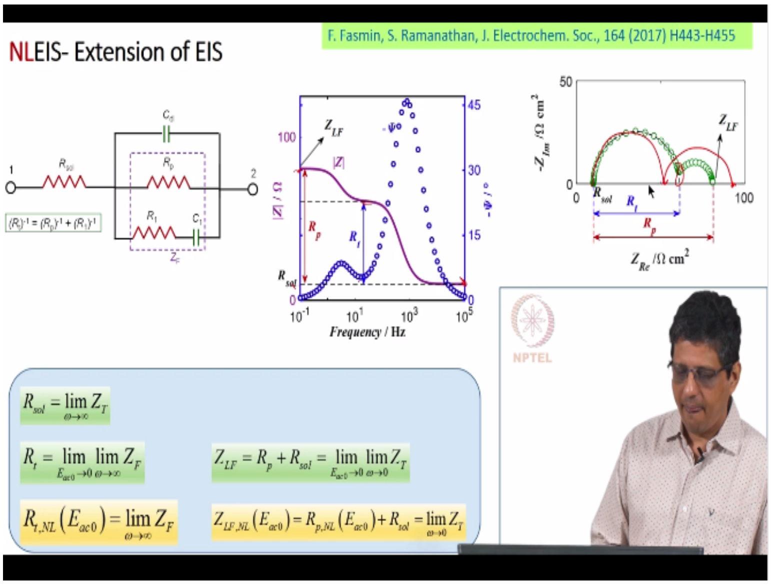

If the perturbation is large, you can still get the data. So what you have seen earlier is complex plane plot, this is just a representation in Bode plot to say high frequency limit is going to give us the solution resistance, the low frequency limit is going to give us solution resistance plus polarization resistance and the next saturation point here which corresponds to the point here tells us that sum of charge transferred resistance and solution resistance.

This could be modeled by a circuit like this. You can also model by other equivalent circuits and get the expression for polarization and charge transfer resistance. Now if I apply a large amplitude perturbation, this spectrum may change and become for example like this. I can still call this as polarization resistance and this as charge transfer resistance except that I have to denote that they are nonlinear values and they depend on Eac0.

So in that case, we drop the requirement that the Eac0 should be small. Eac0 can be large, can be small in general Rt and Rp can depend on Eac0 and we would describe those Rt as Rt, NL and Rp as Rp, NL okay. So at the fundamental as we mentioned before, it is easy to measure the nonlinear impedance data.

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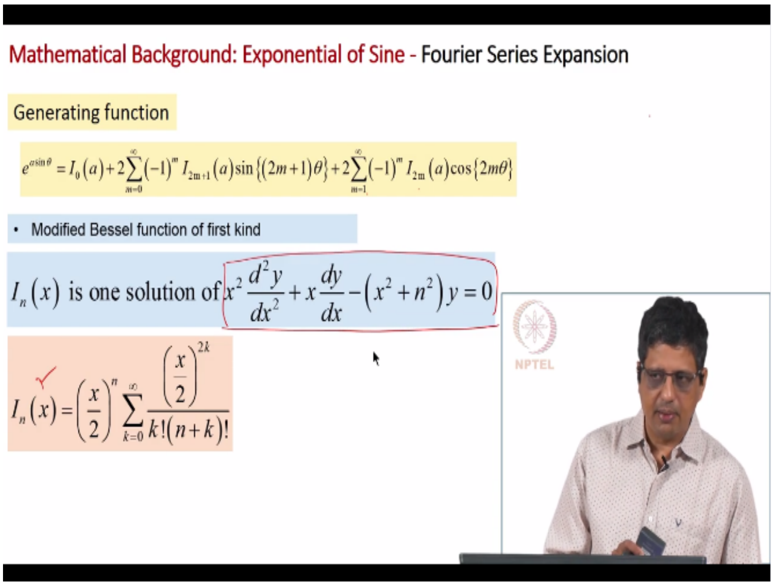

Now I want to give you some background required for getting better handle on this system. One, there are functions called special functions okay. We are familiar with exponential, sinusoidal, cosine, etc but there are many other functions, one of them is called Bessel function and another is called modified Bessel function okay. Modified Bessel function is denoted by In and Kn.

In is basically solution of this differential equation x square y double prime+xy prime-x square+n square y. This case, I have taken n to be integer, it is also possible to get a definition for non-integer values, we are not going to worry about that, we are not going to use them so we are going to worry only about In where n is an integer okay. There are two solutions, this I is called modified Bessel function of the first kind and K is called modified Bessel function of the second kind.

And we would use I and therefore we will restrict ourselves to I. There is a type of function called Generating function. It is relevant for us; therefore, I am describing it here. If you write exponential of a sin theta, it is possible to expand that in Fourier series and that would give us I0 of a that is modified Bessel function of first kind with order 0, the value of a is used here+2 times sigma going from 0 to infinity, Fourier series going from sin omega t's or sin theta, sin 2 theta, sin 3 theta up to infinity.

But first we described only the odd functions, so -1 power m this is again modified Bessel function of order 1, 3, 5, etc multiplied by sin theta, 3 theta, 5 theta, etc. So all the odd functions come in terms of sin, all the even functions are given here, m going from 1 to infinity, just note down, in the previous case m is going from 0 to infinity, it is going from 1 to infinity, it is an index value.

This is an index value, right now we will just focus on this. This comes in the same way except that we get a cosine that means it is a sine wave with pi/2 offset and if I want to get the value of In of x, In of 1, In of 0, In of 0.5, In of -2, any of those values, I have to use this series okay. So it has x/2 power n and then it goes with an index of k going from 0 to infinity, k factorial n+k factorial in the denominator, x/2 whole thing raised to that power 2k in the numerator.

Of course, we do not need to do the summation. If you have access to software like MATLAB, you can just call this Bessel function and in MATLAB this will be called Bessel i n, x. You can give the integer value, maybe n=1, n=2, n=0 and then x whatever value you want to put we can substitute there and similarly in other mathematics software, you would be able to get access to this function. It is just for us to know how to expand this in series and compare things.

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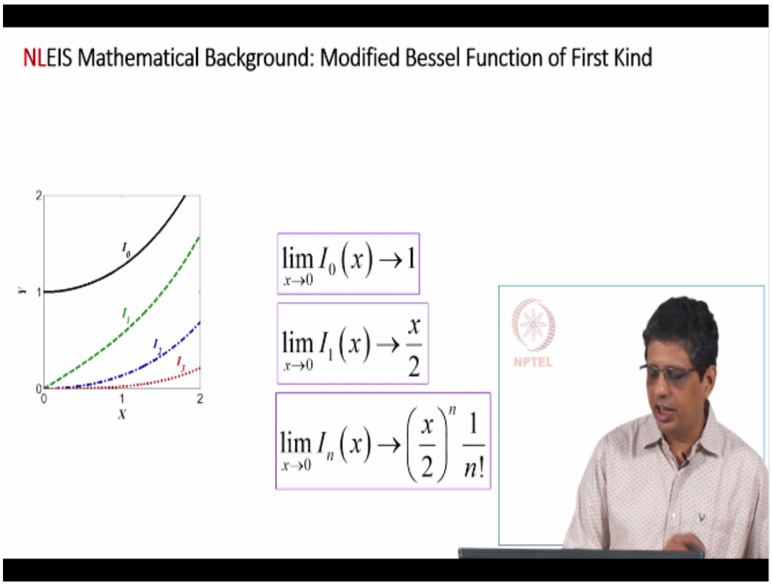

How do they look okay? I0, I1, I2, I3 values for a small range of x value. They look somewhat like exponential functions. They are not exponential functions but they look somewhat similar. I0 when x tends to 0, I0 will tend to 1. Any other modified Bessel function, any other meaning I1, I2, I3 all of them will tend towards 0 at different rates. So limit x tending to 0, I0 x is 1, limit x tending to 0 In in general for integer n.

It is going to be given by 1/n factorial x/2 power n and we will use this information at the next calculation okay. So as x tends towards 0, I1 tends toward 0, I2, I3 they all tend toward 0 but at different rates okay this is one.

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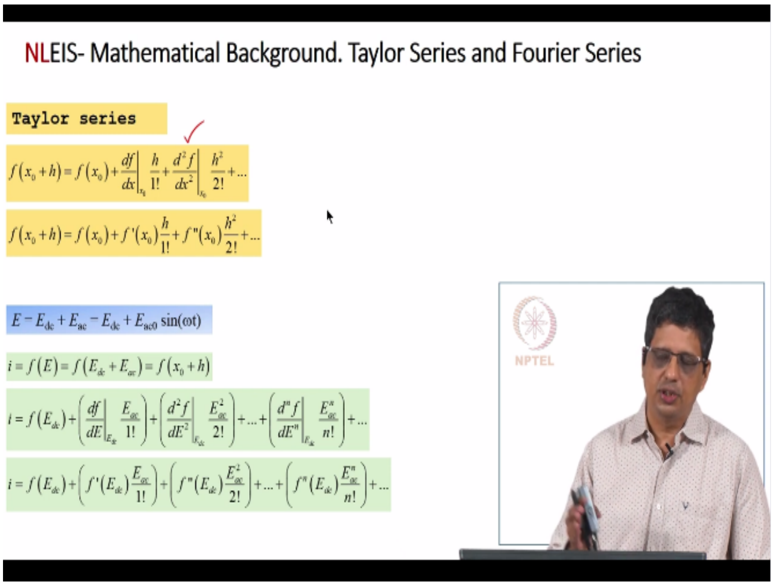

Next, there is a series called Taylor series. If you are given a function f of x, we know the value of the function at a point at x0 and we want to find the value of the function near that point. So we move a small distant h away from the x0, we can write this function in the Taylor series okay. So we would write f of x0+h=f of x0+the first derivative evaluated at x0*h/1 factorial, second derivative evaluated at x0*h square/2 factorial and so on.

And sometimes we use a different notation, we will say f of x0, f prime to indicate first derivative evaluated at x0. So we will call it as f prime x0, f double prime x0 and so on. Now I want to look at the potential, E as Edc+Eac where Eac is given by Eac0 sin omega t. Instead of looking at x0 and h, we will look at E, x0 equivalent will be Edc, h equivalent is Eac. Now I can write current as a function of potential and I would write it as f of Edc+Eac.

That is equivalent to writing it as f of x0+h and you can substitute here, you can say f of Edc df/dE or f prime Edc, f double prime Edc, f nth derivative at Edc. So it is essentially substituting for x0 and h with Edc and Eac and you get a Taylor series expansion. We have not defined what that relationship is.

We have not expanded f of E yet okay. Still I want to go through this for you to get an understanding of how to write the Taylor series expansion and truncate after few terms, not just after the first term.

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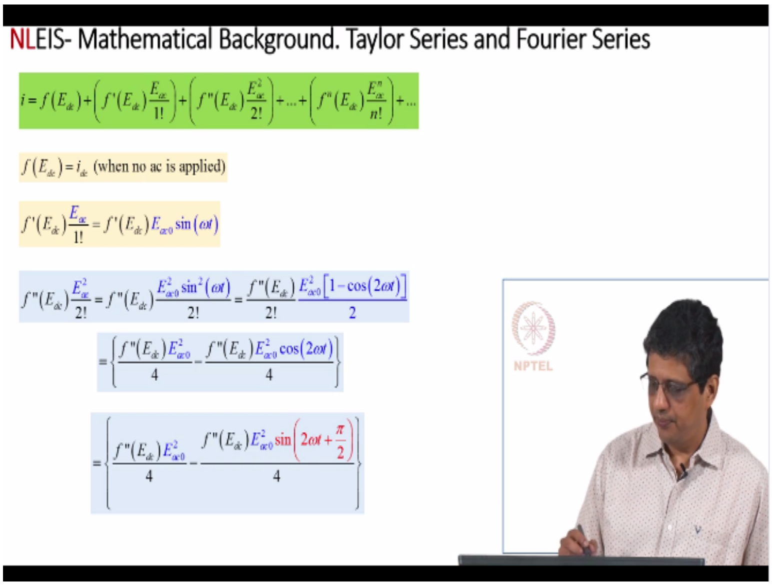

So this is the expression we got in the previous page. If ac is not applied, we apply only a dc potential, we will get a dc current. So that means F of Edc will normally denote as Idc, f prime Edc we keep it as it is, we are just going to keep it as it is, Eac we are going to replace with the ac 0 sin omega t. This is ac, we have done this before. This next term, we are going to keep f double-prime Edc that is second derivative of f with respect to potential evaluated at Edc.

We have the 2 factorial, Eac square we are going to expand this and then we are going to write the trigonometric identity sin square is 1-cos 2 theta/2 which means here it is going to be 1-cos 2 omega t/2. This is going to give us two terms; one is going to be a constant term that is independent of omega, another term which is 2 omega t okay. This cos 2 omega t we can write it as sin 2 omega t+pi/2.

So basically it is a second harmonic with a phase offset. So this should come as f double prime Edc/4 Eac0 square-either cos 2 omega t or as sin 2 omega+pi/2.

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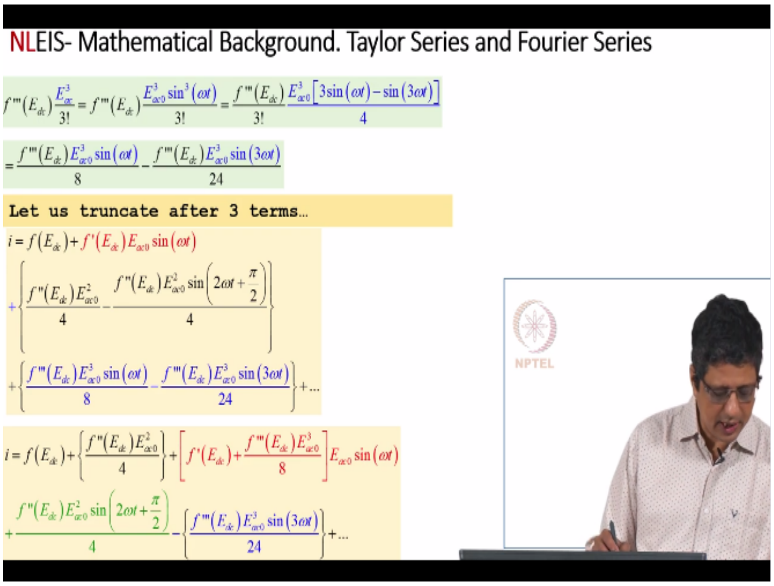

And if we write the third series, third term, sin cube we can write it as 3 sin omega-sin 3 omega which means I can rearrange it. So I will get one response at omega, one response at 3 omega arising out of sin cube. So we will say we are going to use up to 3 terms and then truncate because in Taylor series you can write up to infinite number of terms and then you can rearrange them and group them or we will truncate after 4 terms, 5 terms.

The more number of terms you take; more accurate the calculation is going to be. So if I say I am going to only take up to 3 terms, so you can write first term dc, next term is the first response at fundamental, next term is response at second, next term is the response at sin cube which gives us fundamental as well as 3 and you can rearrange. When you rearrange you see the dc offset has the f Edc, it also has some contribution from the second term.

First harmonic fundamental has contribution from the normal response at sin omega; it also has contribution from the sin cube. Second harmonic actually comes from the sin square, third harmonic comes from sin cube.

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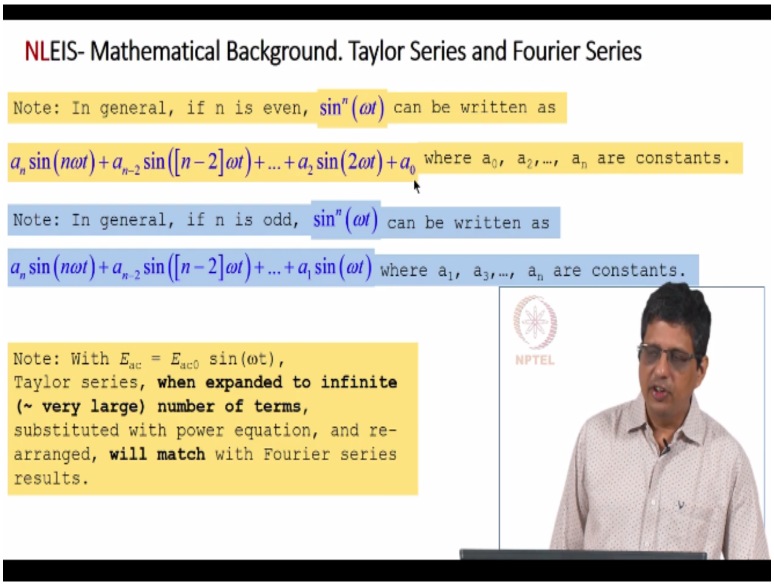

In general, if I write in terms of sin power n omega t, we can write it as a series sin n omega t, sin n-2 etc until we get to a constant. That means if I start with sin power 10 theta, I will get sin 10 theta, sin 8 theta, sin 6 theta, 4, 2 and then a constant. If I start with an odd power okay, if I start with power 7, sin power 7 theta or sin theta raised to the power 7, I would get 7 theta, 5, 3 and 1 okay.

And of course, these values are constants known constant, we just have to go through the trigonometric identities and write this. So that means if I want to know the contribution to sin omega t, I should look at sin cube, sin 5, sin 7 until infinity okay and likewise contribution to the second harmonic will come from sin square, sin power 4, sin power 6, etc. If we say that after certain time, we can truncate then up to that we should count this and rearrange okay.

When we write Eac as Eac0 sin omega t okay, if you write Taylor series up to infinite terms or very large number of terms, substitute with the power equation okay, rearrange, then whatever we get out of that will match with the Fourier series expansion. It is not always possible to write Fourier series expansion for many of the functions, many of the relationship. If you write for infinite number of terms, it will match correctly.

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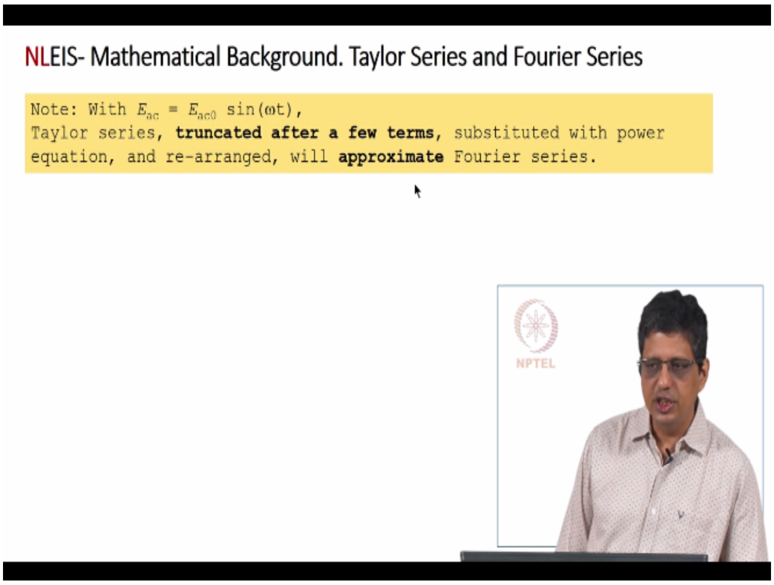

If you write for few terms, truncate after some term, it will be an approximation. So Taylor series truncated after a few terms and then substituted with power equation, trigonometric identities, rearranged, will approximate each coefficient.

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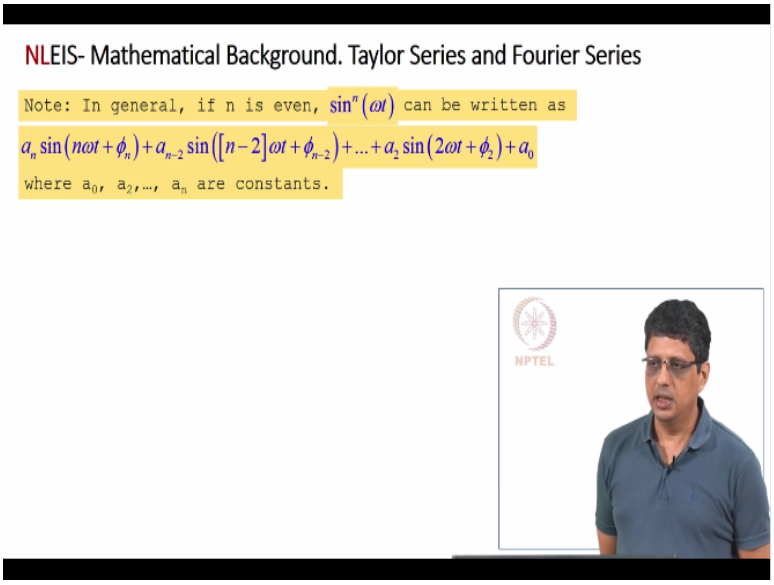

So to complete the information on the mathematical background, we have seen Taylor series expansion and we want to compare with the Fourier series expansion. So in general if I have a Taylor series expansion, I would have Eac sin omega t power 1, power 2, power 3 in general nth power and when I write nth power that is sin omega t raised to the power n, it can be written in terms of Fourier series.

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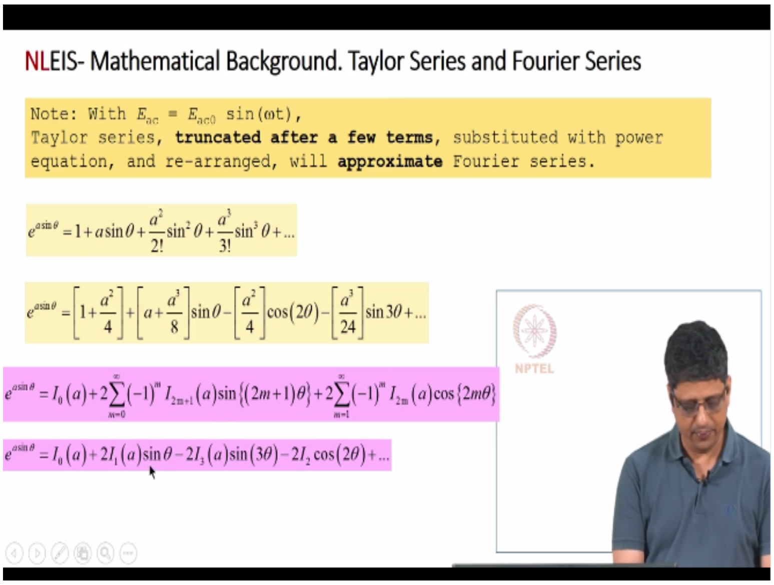

So if it is Taylor series truncated after few terms and then substituted with the power law and then rearranged, what you would get is an approximation for Fourier series. Take an example of exponential a sin theta. I can write it in Taylor series as 1, so if I write e power x I would write it as 1+x+x square/2 factorial x cube/3 factorial and so on. Here I have truncated it after 4 terms, so I have 1+a sin theta a square sin square theta a cube/3 factorial sin cube theta.

And we have seen that this can be written in terms of cosine and sine with sin 2 omega t or cosine 2 omega t and if I rearrange it, I would get constant the first term in the Fourier series as 1+a square/4. This next term is sin theta, the coefficient is going to be a+a cube/8 because sin omega t or sin theta raised to the power 3 will come sin 3 theta and sin theta with the corresponding coefficient.

Then, sin theta is raised to the power 2 will give me cos 2 theta, so I have coefficient of -a square/4 and of course the 3 theta comes from the sin cube, it is a cube/24. This is basically approximation for the Fourier series coefficient. We also know the exact Fourier series for this, e power a sin theta it can be written in terms of modified Bessel functions and we have seen that all the odd theta values will come in terms of sine and all the even thetas will come in terms of cosine.

So we can take this series, we can stop at sin 3 theta that is it will have a constant, sin theta, sin 2 theta and 3 theta. So I am going to truncate it at that, so in this case I0 is the one corresponding to the constant. If I substitute m=1, I would get sin theta, I am sorry. If I substitute m=0, I will get sin theta, m=1 will give a sin 3 theta. In the second part of this, if we substitute m=1, we will get cosine of 2 theta, m=2 will give me 4 theta.

Since we are going to truncate after 3 terms we will just stop at I3, so I have I0, I1, I2 and I3 and then of course you have negative value for I2, negative value for I3, positive value for I1.

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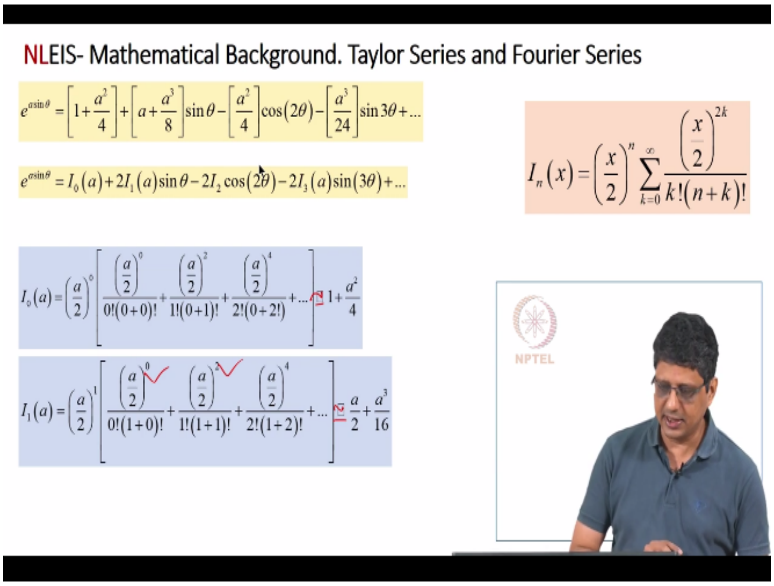

This is Taylor series expansion truncated and rearranged. This is Fourier series expansion truncated after sin 3 theta and of course for the each modified Bessel function, we know how this can be expanded in the power law power series. So for example, if I want to calculate I0 of a, it can be written as a/2 raised to power 0 and then summation with 0 to 4 and so on. This is approximately equal to 1+a square/4 when we stop at a cube.

So this is going to give us constant, this is going to give us power 2, this going to give us power 4, we are going to neglect anything more than power 3. Therefore, we are going to write it as 1+a square/4. I1, this is a/2*a constant a squared term a power 4 term. We are going to neglect this. So we are going to take only this term and this term and we will write approximately this is a/2 and a cube/16.

Now you have to remember I0 term is corresponding to 1+a square/4 this comes correctly. I1 term has to be multiplied by 2 and then if you see compare this Taylor series and the Fourier series, you will see that they are matching when this is truncated after a cube term and I2 term, correspondingly if you do this you would see that it is matching correctly and I3 also when you truncate it, it will match correctly with the Taylor series expansion.