Ultrafast Optics and Spectroscopy
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Lecture - 13

Welcome back to the module 3, we are continuing non-linear effects which are generated due to the propagation of ultrafast pulse through the dielectric medium.

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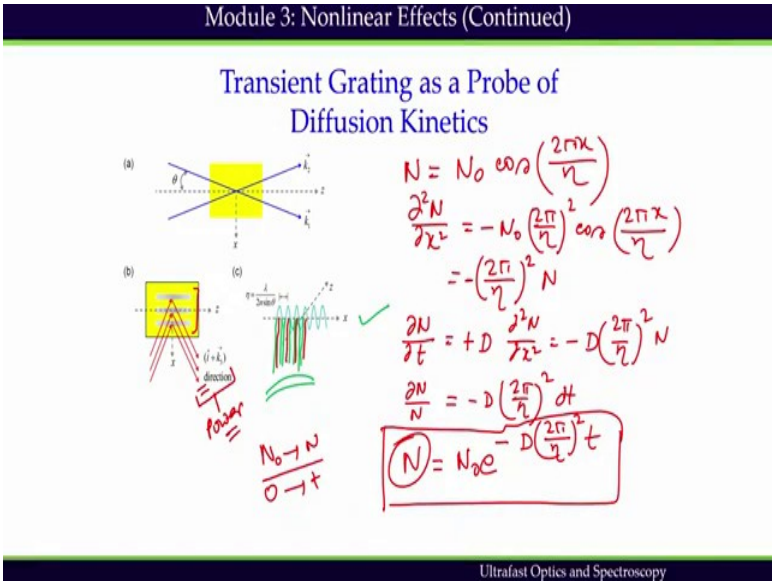

So, far we have discussed the many non-linear effects. And in the previous lecture we have discussed third order non-linear effect including transient grating and polarization grating those are third order processes.

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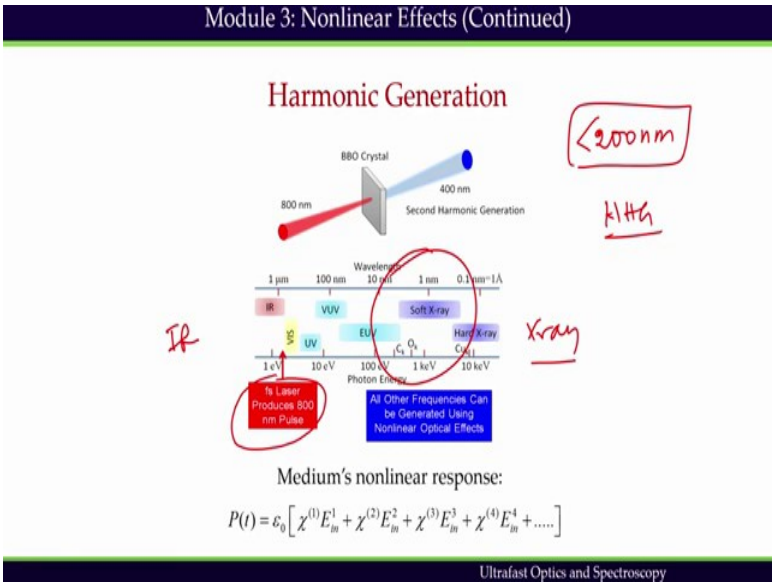

Now, we will discuss higher order process. Higher order process including high harmonic generation, most of the ultrafast laser sources synthesize ultrafast pulses with center wavelength at 800 nanometer. This is we have already shown that the laser sources primarily depends on the Ti sapphire crystal. And Ti sapphire crystal can gives the emission at 800 nanometer, near 800 nanometer that is why most of the laser system ultrafast laser system produces this kind of 800 nanometer pulses.

We have seen that non-linear optical techniques play a pivotal role to further up shift or downshift light from 800 nanometer to different wavelengths. It can be like we can go to IR regime, we can go to X ray regime with this non-linear frequency conversion processes. Non-linear frequency conversion techniques almost exclusively rely on crystalline solids as non-linear medium. We have already seen that for efficient conversion phase matching must be achieved and this can be achieved by relying on an isotropic material birefringent crystal thickness also plays an important role.

The reliance on solid state materials severely limits the application of non-linear optical techniques to very short wavelength. We cannot create short wavelength, very short wavelength less than 200 nanometer, we cannot create in a crystal because crystal is start absorbing those high energy photons. The best solid non-linear material such as lanthanum, boron oxide and barium boron oxide can be used to generate light at wavelengths as short as 200 nanometer.

Most of the solids are not transparent at wavelengths shorter than this, non are transparent in the extreme ultraviolet or X ray regime of the electromagnetic spectrum. On the contrary many gases are transparent well into vacuum ultraviolet and even shorter wavelengths can propagate through the medium through the gas medium with moderate absorption. This is why one can think of performing high harmonic conversion of 800 nanometer light into EUV extreme ultraviolet regime.

Regimes are shown here in a diluted gas medium. We shall see that High Harmonic Generation HHG in short is called HHG, this high harmony generation in dilated gas medium provides a useful source of XUV or soft extreme.

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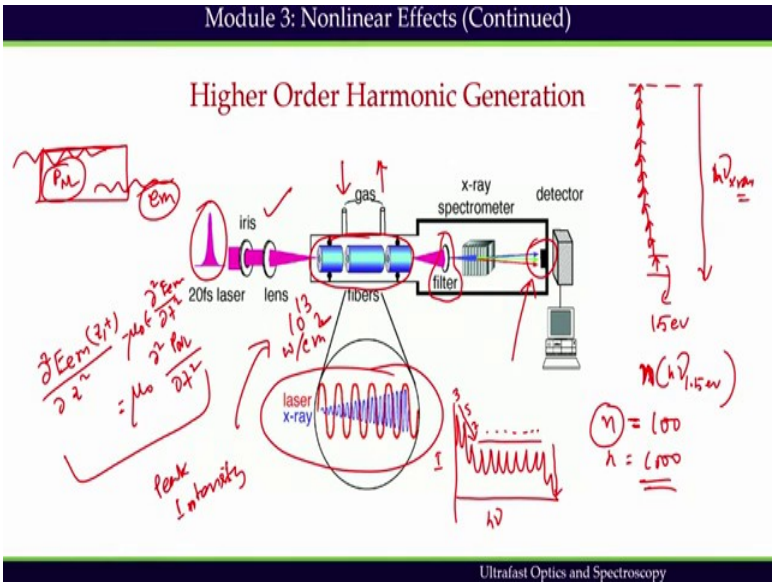

High harmonic generation represents an extreme non-linear process, in which more than 10 or even 100 input photons from a visible or near infrared laser are combined together in a frequency of conversion process. One can say that I have this all this photons clubbed together of 800 nanometer, 800 nanometer is approximately 1.5 ev.

So, all these photons will be clubbed together and I can create a big photon that is in x ray regime. And this number can go up to hυ is 1.5, n number of photons which can be clubbed together they can be of the order of let us say 100, this n can be 100, it n can be even 1000.

So, this is an extreme non-linear process where it is not third order, fourth order it is going to be 100th order or 2000 order. So, this kind of high order harmonics can be generated with the help of high harmonic generation process. HHG provides high harmonic generation provides an attractive source of ultrafast pulses in the extreme ultraviolet and soft x ray regime. When we derived 1 D equation one dimensional equation for non-linear optics, if we remember this was the equation which we have driven.

So, basically what was the picture we had in the derivation of the in the non-linear propagation of the ultrafast pulses? We said that there is an input beam then that is creating non-linear polarization and that non-linear polarization is created emitted beam. Now, question is how this induced polarization and emitted beam they are they are connected; they are connected with the help of this equation this is Zt /Z 2 minus μ naught epsilon E this is emitted beam, 2 equals mu naught 2 P non-linear polarization d 2 t.

So, this was the equation we got as a non-linear equation of optics. And this equation was obtained assuming that the non-linear polarization is weak, that is the perturbative limit. So, he is assumed fairly weak non-linear process that is the perturbative limit is employed which explained the numerous non-linear optical phenomena already. We have seen that phase matching particularly is one important factor which we have got from this equation from if the conditions was obtained from this equation. And, second harmonic generation we have seen self phase modulation we have seen from this non-linear optical processes.

But those processes are lower order processes for HHG process this perturbative limit breaks down because of the extreme non-linear nature of the process. As I have told you that it can be 100th harmonic we are generating. HHG occurs when an intense ultrafast pulse is focused into a noble gas medium such as helium, argon etcetera as depicted here. So, I have a gas inlet and outlet. So, it is going inside and I have femtosecond laser pulses focused inside this gas medium this can be fiber, this can be optical fiber or this can be gas jet or this can be gas cell.

Once we focus that beam then we create the x ray in the at the focal point only at the focal point and focal region and then we can filter the fundamental beam. We can use

metallic filter because we know that high frequency photons having higher penetration depth as compared to the low frequency photon. So, that is why we can use a aluminum filter or metallic filter and then if we disperse the beam we will be able to see the spectrum. And generally the spectrum how does this spectrum look like high harmony generation spectrum, it typically it looks like this, this is the hυ photon and this is the let us say intensity.

It creates third order, then fifth order, then probably seventh order, then there is a Plato and then suddenly there will be cut off. So, this looks like this there is a Plato regime, there is a lower order harmonics gradually decreasing quickly that is third, fifth and always odd harmonics will be produced.

So, this is your third, then this is fifth, this is seventh then so on blah blah and then suddenly there will be a sharp cutoff in the spectrum. So, that is the very common feature of the HHG spectrum which we can monitor. The required focus intensity of the fundamental beam, this fundamental beam focus intensity has to be at least 10 to the power 13 watt per centimeter square.

Meaning of this 10 to the power 13 watt per centimeter square is now obvious what does it mean we know that. And, this is the peak intensity and we needs very high peak intensity to obtain this high harmonic generation.

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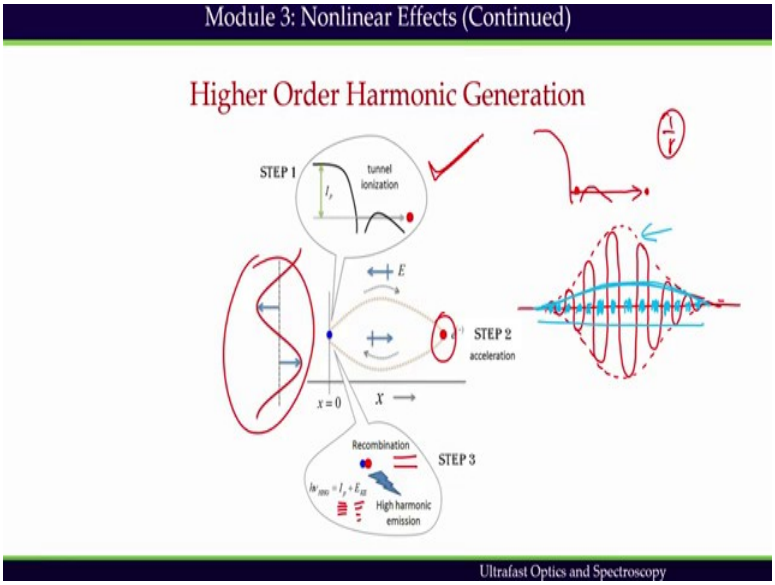

Although the most accurate description of HHG process involves numerical solution of the non-linear Schrodinger equation a quasi classicals three step model proposed by Corkum can also accurately predict the general feature of HHG. According to this model as depicted here, in the first step the intense laser field suppresses the coulomb potential of the atom.

So, this is the first step when I have electron attached, so, will express it let us say I have a nuclei and electron sitting together. Now due to interaction of the strong laser field, laser field is lesser propagating along this direction. And when laser field is propagating along this direction the field is changing its polarity along this direction is perpendicular direction and in every half cycle is changing the direction.

So, what will happen this field will exert a force on the electrons. So, once tunnel ionization occurs due to strong field this electron is free and this electron is experiencing the electromagnetic wave which is propagating along through the medium. So now, what will happen due to this field at a particular point this electron will be accelerated away from the atom the a parent ion, cation. And, then after a certain time which is half cycle of the optical cycle after the optical half optical cycle this electron will come back again because the polarity has change and it will come back.

And there is a possibility the probability is very low, but there is a possibility that is the electron will find it is own parent cation or parent ion and it will be combine and when it will recombine it will give the emission. So, that is all about high harmonic generation process is a three step process one can explain. The first step is tunnel ionization which will make the electron free then electron will propagate along in the field and that will acquire kinetic energy and then they will recombine. This recombination step is the last step, when they will recombine they will emit the field and when their emission occurs they will emit the ionization potential plus the kinetic energy it has gained, when it was traveling in the field.

So, the first step; in the first step the intense laser field suppresses the coulomb potential of the atom or the molecule allowing tunnel ionization the tunnel inhalation process is something like this. So, I have a coulomb potential which is something like 1 by r that is the Coulombic potential and I have an electron here. So, due to strong field what might happen this potential will be suppressed down to this and suddenly this electron can be

tunneled out and I can have this electron free electron that is called tunnel ionization process which is the first step of HHG process.

Second step involves evolution of the free electron in the laser field. As the optical field oscillates, it first propels the electron away from the ion and then when the field reverses the field accelerates the electron back towards the ion. Finally, the third step is recombination of the free electron with this parent ion emitting high harmonic photon. The ionization and recombination can occur in vey every half cycle of the driving field. So, if I consider the driving field instead of this one of cycle I can now consider, we know that a pulse may have a number of cycles and I can represent it like this way.

So, this is called carrier wave of the pulse and it has a number of cycles and in every half cycles I will produce this HHG which means that here I will produce HHG, here I will produce HHG, here I will produce HHG, here I will produce HHG and continuously I will produce HHG, HHG pulses ok. Each pulse is of the attosecond time scale, of the attosecond duration, but overall this pulse would be femtosecond just like the parent beam.

So, in every optical half cycle I will have a burst of electromagnetic energy. This is called the attosecond pulse train, but the overall total pulse duration of the XUV beam is going to be femtosecond or same almost same as the pulse duration of the driving laser field

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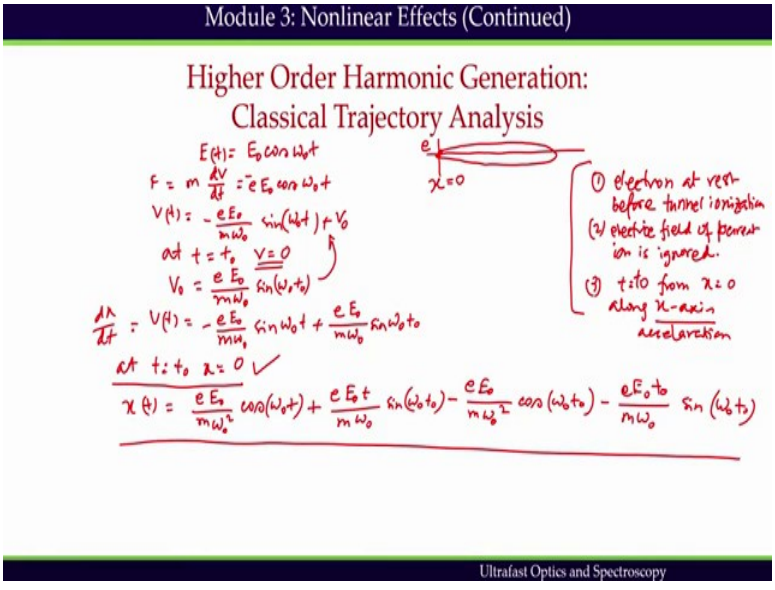

This three step model can be expressed analytically and we will look at the analytical expression. The real field is represented by let us say E t equals E naught cos omega naught t, I will consider this one. We are not considering pulse here because it is the each optical cycle which is important that is why we are considering only the plane wave expression. Here we have considered that feel does not depend on space coordinate.

This is reasonable approximation as long as wavelength of the input beam is large compared to the atomic size. In addition a few assumptions are employed electron was at rest in an atom before tunnel ionization. So, this is the process we are considering we are considering that it is coming back to x equals 0 starting from x equals 0 it is moving away and then coming back to x equals 0 that is the trajectory of the electron.

Electron was at rest in the atom before tunnel ionization, electron was suddenly completely free. So, there are few assumption we have made here is that, electron at rest before tunnel ionization. Second assumption is that electron was suddenly completely free, electron will face only electric field of the laser electric field of the parent ion is ignored. The electric field after we have ionized it electric field of parent ion is ignored and electron is accelerated at time t equals 0, t equals t naught from position x equals 0 along x axis this is the acceleration.

So, this is the assumptions, we have made under this assumption the force experienced by the electron due to oscillating electric field is given by. So, I had an electron here at this point and I want to know the force. So, force is going to be m dv/dt which is nothing, but minus e E naught cos omega naught t. So, if we have this then we can integrate it and we can get the velocity to be minus e E naught by m omega naught sin omega naught t plus V naught this is the velocity we get, according to the assumption made above electrons was at rest at the time of ionization.

So, we can say that at t equals t naught v equals 0, I can say that. And if we have this initial condition then we have V naught equals e E naught by m omega naught sin omega naught t naught. So, we can insert that here and we get V t is nothing, but minus e E naught by m. We get V t equals minus e E naught by m omega naught sin omega naught t naught sin omega naught t plus e E naught by m omega naught sin omega naught t naught.

Similarly, we can integrate with respect to now we can we can express this one as dx dt and we can integrate. And when we integrate it within this limit at t equals t naught we have x equals 0 and that is the initial condition we this is the boundary condition we employ. Then we get x t value also which is can be expressed as e E naught divided by m omega naught square cos omega naught t plus e E naught t divided by m omega naught sin omega naught t naught minus e E naught divided by m omega naught square cos omega naught t naught minus e E naught t naught divided by m omega naught sin omega naught t naught. This equation features the oscillatory trajectory of the electron in the input field.

Now, this is the expression for the trajectory of the electron which will follow under the assumption of following assumption. Assumption is that force assumption is at t equals t naught at the initial time x equals 0 that is the we started with x equals 0 and at t equals t naught velocity was also 0, that is also one more assumption we have taken.

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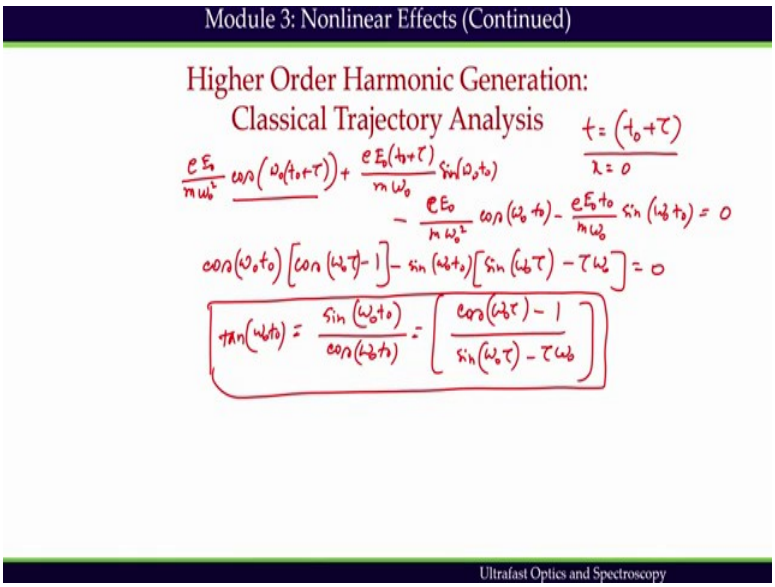

And so, with this assumption we get an expression for x it is also assumed that at the time of recombination electron comes back to rest; if time taken by electron for acceleration and recombination is defined by tau(τ). So, if I express tau as t naught sorry t that is the time in it this time of recombination if I express as t naught plus tau then and this recombination occurs when x equals 0 then we can write down that, we can write down following expression e E naught divided by m omega naught square cos omega

naught t naught plus tau plus e E naught t naught plus tau divided by m omega naught sin omega naught t naught minus e E naught divided by m omega naught square cos omega naught t naught minus e E naught t naught divided by m omega naught sin omega naught t naught equals 0.

So, we can now use this cos a plus b identity, this can be decomposed cos a plus b equals cos a cos b minus sin a sin b. We can use this identity and we can decompose it and if we decompose it we will be able to cancel out few terms and then we get this. We get following expression one can do this derivation very easily. Cos I am going to write down this final expression, cos omega naught t naught multiplied by cos omega naught tau minus 1 minus sin omega naught t naught multiplied by sin omega naught tau minus tau omega naught equals 0 we get this expression.

Or we can write down this sin omega naught t naught divided by cos omega naught t naught equals cos omega naught tau minus 1 divided by sin omega naught tau minus tau omega naught, this is the expression we get. Or this can be written as tan omega naught t naught equals this. So, this will be useful this expression will be useful to further reduce the expression, the kinetic energy expression which we will write down in the next slide.

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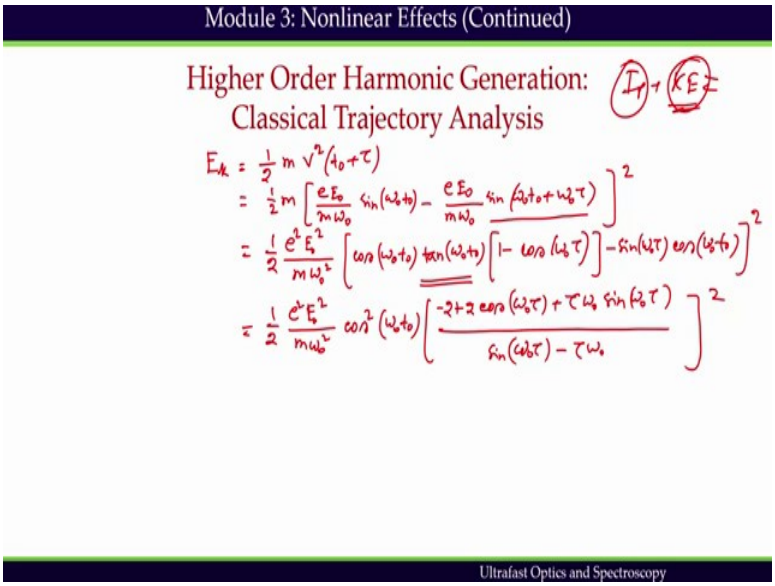

Now, we shall go back to the equation which features electron trajectory to estimate final kinetic energy. Because remember we are interested in I p plus kinetic energy and we are interested in knowing the kinetic energy gained due to the external the electromagnetic

field we have. I p is constant for a system I p is dimension potential, that is constant we cannot change it for a system. But KE should depend on the external electric field and that is exactly what we are trying to calculate.

So, kinetic energy E kinetic energy is can be expressed as half m v square and v has an expression v is a function of t naught plus tau. So, at the time of recombination the time is t naught plus tau, I have to find out the kinetic energy and that kinetic energy can be expressed as half m then I insert the expression for v that is e E naught divided by m omega naught then sin omega naught t naught minus e E naught divided by m omega naught then a sin omega naught t naught plus omega naught tau whole square this was the expression for v at the time of recombination.

So, we can simplify this one further and we can get this expression and we can actually decompose this sin function as was sin a cos a plus sin a cos b. So, this trigonometric expression we can decompose and finally, I am writing the final expression which we get is half then e square E naught square divided by m omega naught square multiplied by cos omega naught t naught multiplied by tan omega naught t naught then multiplied by 1 minus cos omega naught tau minus sin omega naught tau cos omega naught t naught whole square. So, this is the expression we get.

And we remember tan omega naught t naught we got an expression previous in the previous slide. So, we should plug that in, the moment we plug that in finally, we get this expression and we can simplify this expression further to get this expression half e square E naught square divided by m omega naught square then cos square omega naught t naught multiplied by minus 2 minus plus 2 cos omega naught tau plus tau omega naught sin omega naught tau divided by sin omega naught tau minus tau omega naught whole square, we get this expression.

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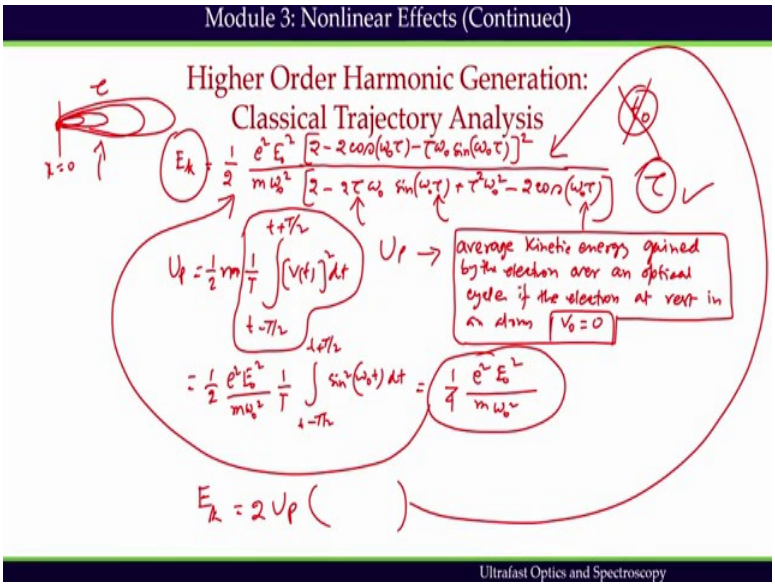

And, if we perform trigonometric identities you can use trigonometric identities to write down this final expression for E k that is the kinetic energy one can do this math very easily half e square E naught square divided by m omega naught square then 2 minus 2 cos omega naught tau minus tau omega naught then sin omega naught tau square divided by 2 minus 2 tau omega naught sin omega naught tau plus tau square omega naught square minus 2 cos omega naught tau. So, this is the expression we get, final expression for the kinetic energy at the time of recombination.

So, this kinetic energy should be added to the photon energy, emitted photon energy. Thus the above equation suggests that the final kinetic energy of the electron due to acceleration and recombination depends on total time taken due to acceleration and recombination. Now let us, so, what we see here is that this expression depends only on tau. Tau is the time taken total time taken for the acceleration and recombination. We do not have any t naught component in this expression, we have with this expression depends only on tau that is the total time taken for this.

So, basically if I, at this point if I remove this is actually x equals 0. If I remove an electron this time total time is tau and final kinetic energy depends on this total time. What is the trajectory, if the trajectory is like this, is fine trajectory can, so, different trajectory I can have. Although I am showing it with the lobe, but it is a one dimensional problem we have solved. So, it is actually are going along x direction just, but for visual

clarity I I am showing it through the lobe, showing the trajectory going away and then coming back.

Now we will derive an expression for the ponderomotive energy that is Up which is defined as the average kinetic energy. That is Up now I will define, that is defined as average kinetic energy gained by the electron, if the electron this is gained over an optical cycle if the electron was assumed to be at rest in an atom that is we will consider V naught equals 0.

So, this is what we will consider if V naught equals 0 in the previous expression we have not said V naught to be 0. We said that initially we can have a velocity, but if the V naught is 0 then the average kinetic energy will be considered to be ponderomotive energy and in that case ponderomotive energy U p can be expressed as half m half m 1 by capital T is the period integration is going to be t minus T by 2 from t plus T by 2 this is the integration limit V t whole square d t this is the integration we want.

So, this 1 by T term this is the average quantity determine to determine average quantity over an optical cycle. This is the way we do this. And if we insert the V t expression without V naught term if the V naught term is removed then I get only sin function and I can write down this as half e square E naught square divided by m omega naught square 1 by T then t minus T by 2.

And t plus T by 2 sin square omega naught t dt which is nothing, but 1 by 4 square E naught square divided by m omega naught square. This is the and the average of sin square function over an optical cycle is going to be half. That is why we can take this integration as the we can get the value of like this 1 by 4 e square E naught square divided by m omega naught square.

So, now if we see that part of this expression is here, is present here in this. So, we can plug that in and we can get that kinetic energy expression E k final kinetic energy expression E k equals 2 Up ponderomotive energy multiplied by whatever we have written here, this expression whatever we have written here. So, 2 Up multiplied by this.

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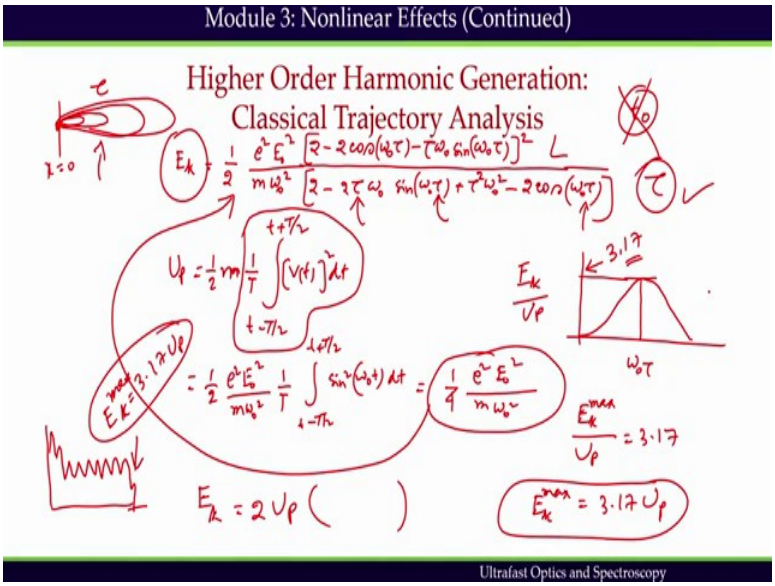

So, if we plot this Ek by Up, then as a function of omega naught tau if you plot that then we get an expression, we can we will get a plot like this which will having a maxima. And, this maxima will appear at the value of 3.17 this is a characteristic number and that is why we can say that E kinetic energy maximum divided by U p is going to be 3.17. Or in other words we can write down that E kinetic energy maximum equals 3.17 into Up average kinetic energy gained in the acceleration process.(see the equation in the above slide)

So, I get a maximum value of the kinetic energy, it cannot go beyond that. So, the spectrum which we have previously proposed that the spectrum looks like the an odd harmonics coming down and then there is a Plato and then suddenly there is a sharp cutoff, this cutoff will be controlled by this E maximum kinetic energy which is nothing, but 3.17 Up.

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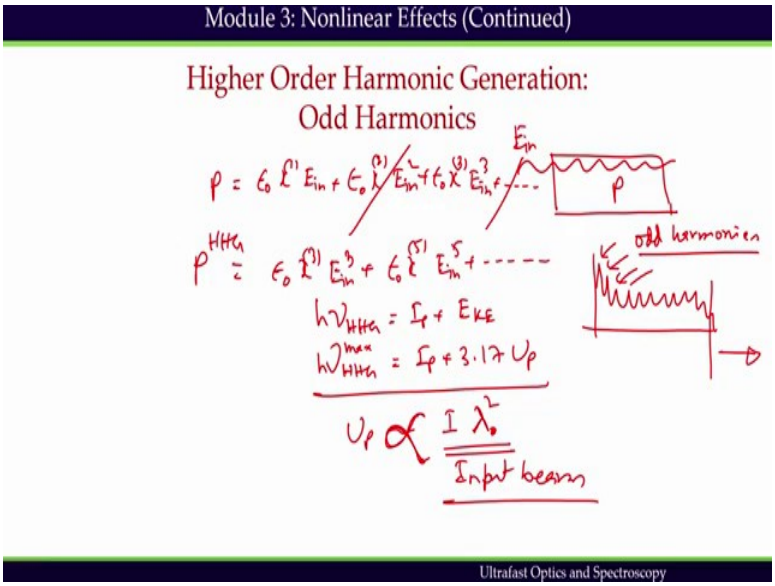

As high harmonic generation occurs in gas medium which features an isotropic medium, and has inversion symmetry the Pe function introduced early in this chapter gets all gets odd symmetry that P function that the polarization. So, we say that we have a medium and the polarization we create due to the input beam propagation through the medium, this is input beam, this is polarization and we said that polarization has this form epsilon naught 1 E input plus epsilon naught 2 input square plus epsilon naught 3 input cube plus blah blah like this way we said that.

Now, because the medium, we are using is a gas phase medium, is an isotropic medium it has an inversion symmetry. So, you know to obtain this inversion symmetry in this Pe function I have to neglect this even terms. So, in the end due to this medium, in the where the non-linear polarization is created for the high harmonic regeneration. So, P^{HHG} polarization will depend only on the odd functions that is E cube plus epsilon naught 5 E to the power in 5 plus blah blah all odd harmonics will be created.

So, that is why when you plot the frequencies we see that this is the spectrum we see generally. And in the and then these peaks are to representing odd harmonics only. So, odd harmonics is generated due to the medium characteristics. With the hυ HHG is nothing, but Ip plus kinetic energy or maximum hυ we have also seen there is a maximum of hυ which can be defined with respect to the maximum the ponderomotive energy which is nothing, but I p plus 3.17 U p that is the maximum energy we get.

And if we want to extend this cutoff, if we want to extend to the higher frequency regime then what we need to do? We need to change the ponderomotive energy. Ponderomotive energy will depend on I intensity as well as lambda naught square. So, they this is proportional to the intensity and lambda naught square. So, both can be changed. These are the characteristics of the input beam. So, for the input beam if we increase the intensity, the ponderomotive energy can increase and the cutoff can increase. On the other hand for the input beam for increase lambda naught then also we can increase the cutoff. These are the very important characteristics of the HHG process.

We will stop here and we will continue this HHG discussion in the next lecture.