Ultrafast Optics and Spectroscopy

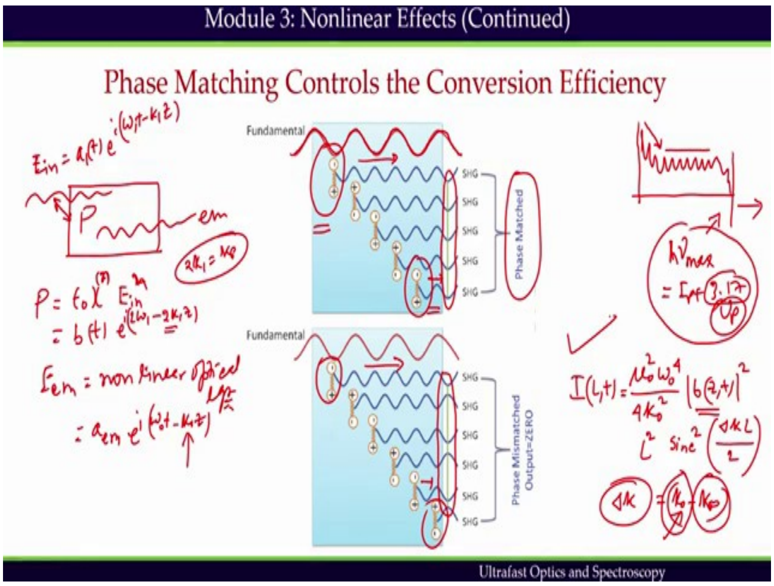
Dr. Atanu Bhattacharya
Department of Inorganic and Physical Chemistry
Indian Institute of Science, Bengaluru

Lecture - 14

Welcome back to the module 3 where we are discussing non-linear optical effects. We in the last lecture, we have discussed high harmonic generation process and so far we have discussed a single atom response. We will recapitulate what we discussed, we had a molecule or atom and intense laser field is propagating along this direction. So, I have considered one atom and then due to tunnel ionization due to this intense laser beam, I have removed an electron from this atom this electron is now free.

And, because the electric field is changing along this direction in every optical half cycle, this electron will face and force due to the electric field and it will go away from the cation and then come back and there is a possibility that this electron will recombine. If this electron is recombining then it can produce high harmonic emission. So, that is the single electron single atomic response or single molecular response we have discussed, but we know that for any non-linear process, phase matching is an important concept.

(Refer Slide Time: 01:41)



So, high harmonic generation will have the same issue and we should understand the phase matching condition which can control the conversion efficiency. Studying the

HHG process in a single atom response model basically provides the predicted shape of the harmonic spectrum. So, what we have seen that that single atom response that three step model can provide us to explain why the cut offs are like this. So, generally this is the nature of the spectrum we see, we see a gradual drop for the lower harmonics third fifth or so on and then there is a plato regime for the high harmonics and then suddenly there will be a cut off and this cut off we have explained with the help of three step model.

We have got the analytical expression for this cut off that cut off we have expressed as $h\upsilon_{max}$ that is going to be Ip plus 3.17 U p and we have seen why we get this 3.17 factor, this is the maximum cut off energy and in order to increase the cut off further to the higher photon energy, what we need to do? We have to change Up that is ponderomotive energy which will depend on the driving laser wavelength as well as the intensity.

But then question is phase matching is an important concept and what does it mean by phase matching? Phase matching is something which we have explained already let us look at second harmonic generation process, we have a fundamental beam propagating through the medium and we said that due to propagation of this fundamental beam we create polarization in the medium.

Polarization is nothing, but oscillatory dipole. So, we consider two dipole which we have created, at the beginning of the medium and in the end of the medium, when the fundamental beam is propagating. So, each dipole will produce its own emitted field and that is the SHG field. Now, this dipole will create this SHG field and this dipole will create this SHG field, if these SHG fields are in phase then we call it phase matching is achieved and we get the higher conversion efficiency, we see the new light generation.

On the other hand, if this dipoles are creating fields which are not in phase anymore they are out of phase then even we have this non-linearity present in the medium we will not see any input any conversion of the frequency. So, phase matching is an important issue and we have to follow certain procedure to bring them in phase to improve the conversion efficiency. To obtain strong harmonic emission, to which means that we have to increase the efficiency of the frequency up conversion process in HHG process, phase matching condition also need to be considered.

A full introduction useful introduction to phase matching conditions is already given in our previous lectures with an example of the second harmonic generation which we have also shown here. This highlights that to produce optical harmonics efficiently, the process must be phase matched. That is the fundamental beam or induced polarization and harmonic beam must be brought in phase throughout the non-linear medium.

Furthermore, we have seen that intensity of the harmonic emission is given by this expression I (L t), L is the thickness which is nothing, but mu naught square omega naught to the power 4, this expression we have already obtained in our previous lectures, b z t L square then there is a cardinal sin function we have seen that is depend that depends on delta kL by 2 and this delta kL this delta k is the phase mismatch which is nothing, but k naught minus kp k naught is the outgoing beam.(look the slides for mathematical expressions)

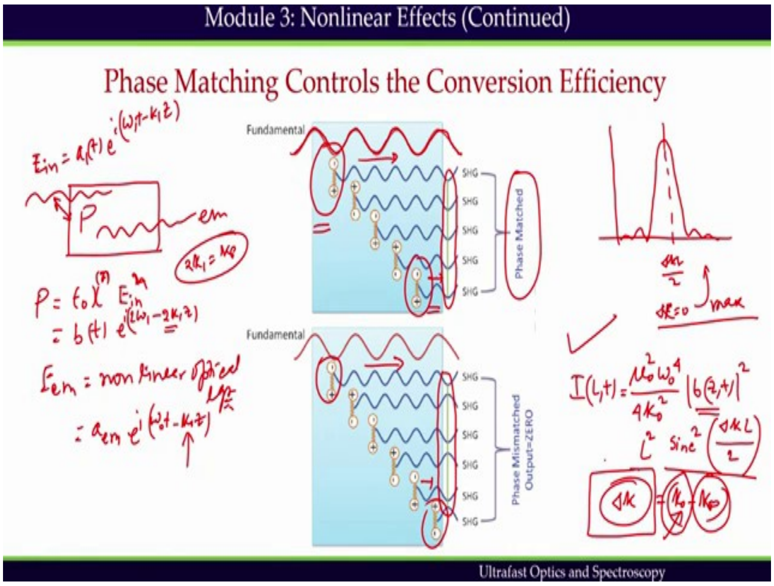
So, basic basically we remind that when we consider input beam this is the input beam, we have expressed input beam as a1 t e to the power i omega 1 t minus k 1 z, this has induced a polarization this polarization will depend on how they are related that they will they are related by the Taylor series expansion. Let say if it is second harmonic generation then we can write down epsilon naught kai second order e to the power input beam square that we have seen if this if this is second harmonic generation. (look the slides for mathematical expressions)

If it is higher order harmonic generation then this is going to be n this will be n n and the this polarization was expressed with the help of b t that is exactly written here b t e to the power i then we said that we will write down this if it is every second order polarization then it is going to be to 2 omega 1 minus 2 k 1 z, it is second order. (look the slides for mathematical expressions)

And so, 2 k 1 2 k 1 is actually kp polarization effecter and then due to this polarization I have created new emitted beam that is E emitted and that is related by the non-linear optical equation, we have written previously and that is that can be written as a emitted beam then e to the power i omega naught t minus k naught z. So, this k naught is the wave vector of the emitted beam and kp is the wave vector of the non-linear polarization. (look the slides for mathematical expressions)

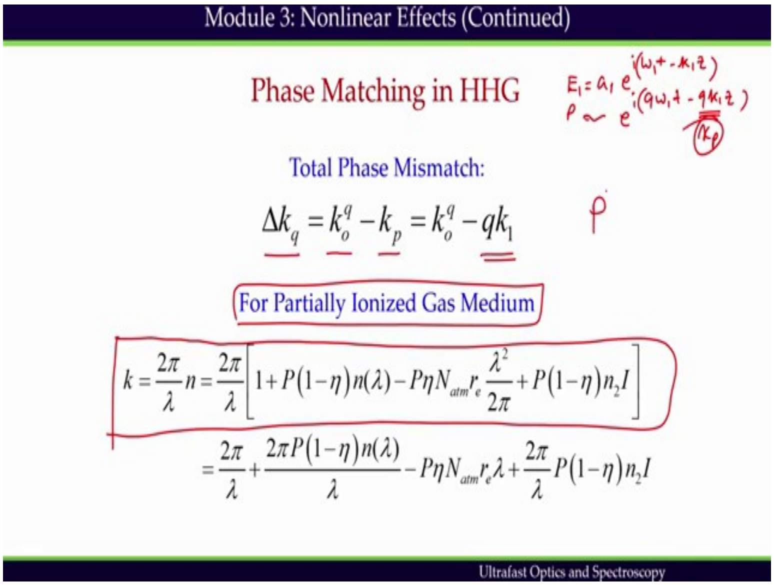
So, these are the expression we have seen and L is the distance travel in the medium, the equation this equation this equation shows that harmonic intensity varies as the cardinal sin functions, this cardinal square sine function this function will control the efficiency and we know that this cardinal sine function if we plot this cardinal sine function then we see that there is a sharp drop which means that little bit of phase mismatch will bring the efficiency down to zero.

(Refer Slide Time: 09:07)



So, this is at 0, if you plot delta k l by 2 then we get. So, cardinal sine functions will be like this, very sharply it drops down its maximum when delta k equals 0, this is maximum and it drops down very quickly when there is a phase mismatch. So, this delta k issue bringing them in phase we have to consider for the HHG process as well. (look the slides for mathematical expressions)

(Refer Slide Time: 09:51)

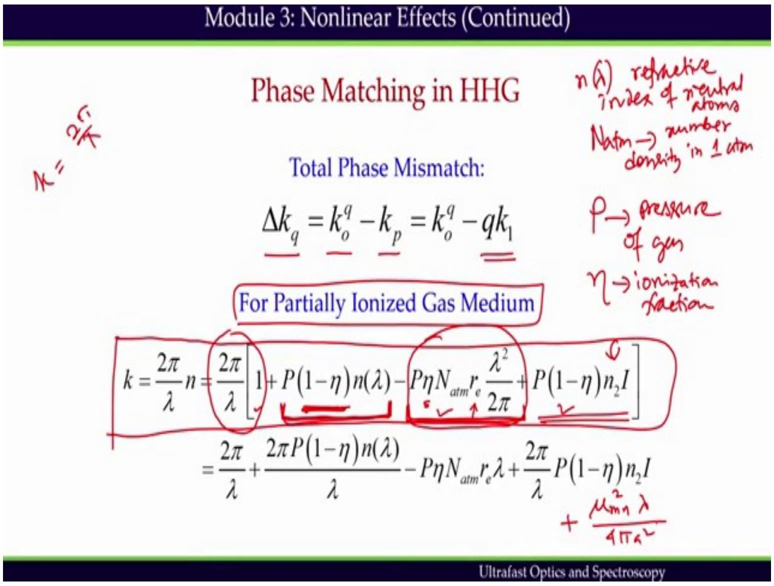


The phase mismatch delta k as mentioned before for the qth harmonics let us say for the HHG process which can be written as delta k q, qth harmonics equals k naught q minus kp which is nothing, but q k 1 k 1 is the input beam, we have shown that it is related like that way because input beam will consider as a 1 e to the power i omega 1 t minus k 1 z that was the input beam. And, the polarization will be then written as if it is qth harmonics then it is going to be e to the power i q omega 1 t minus q k 1 z, this was the expression. So, this is the wave vector of the polarization. (look the slides for mathematical expressions)

So, we have to understand that previously, we considered the refractive index of the birefringent crystal for the second harmonic generation or third harmonic generation, but here we are not using any crystal, we are using partially ionized gas medium. The gas medium which we are using they are partially ionized which means that we have neutral molecules, electrons, cations all are put together.

q is the harmonic order here and k naught q is the wave vector of the qth harmonic k 1 is the wave vector of the fundamental beam and for partially ionized gas medium, the refractive index can be written as this k equals 2 pi by lambda n and n, refractive index of the partially gas medium is given by this where this capital P is the pressure this P here we express in this equation in this refractive index expression. (look the slides for mathematical expressions)

(Refer Slide Time: 11:55)



P is the pressure of the gas medium and eta(χ) is the ionization fraction and n atmospheric it is number density at 1 atmosphere it is the number density in 1 atmospheric pressure and n lambda is the refractive index of neutral atoms. (look the slides for mathematical expressions)

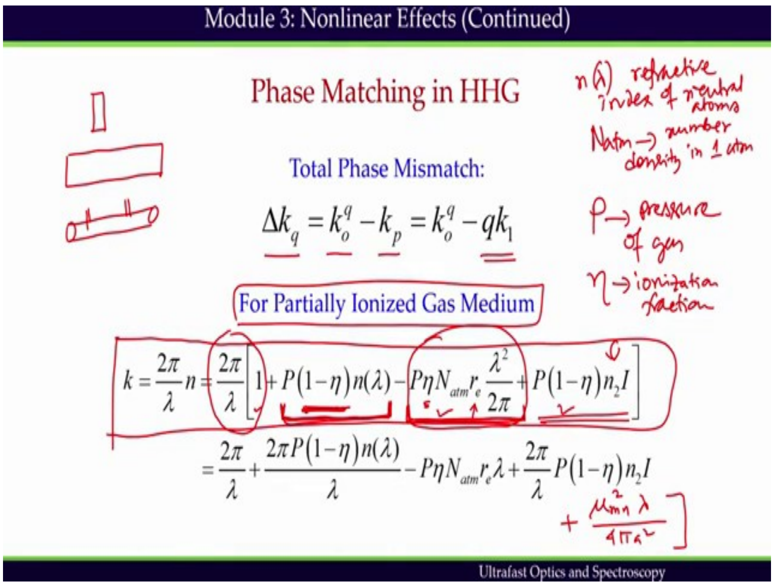
So, what we get is that in a expression, we get we can try to understand the meaning of this expression, there are four terms here one two three and four there are four terms here, the first term contribute to the vacuum dispersion. We know that in vacuum when it propagates we know that it is going to be 2 pi by lambda, that is why the first term is the vacuum propagation. The contribution coming from neutral gas dispersion is coming from this term second term, mutual gas dispersion.

So, 1 minus eta, eta is the ionization fraction which means that 1 minus eta is suggesting that the neutral fraction of molecules which you which you have in the in the gas medium. The third term is the plasma dispersion, plasma is nothing, but combination of it is a fourth state of medium, its combination of electrons ions neutrons everything together has called plasma. So, plasma dispersion is then charged gaseous species and that medium will have a different dispersion that dispersion depends on the pressure then ionization fraction, how much molecules has been ionized and then characteristic radius of electron and then lambda square by 2 pi.

These are the coming from directly from the plasma characteristics of the medium and the last one is the non-linear refractive index which is that is why we have n2 term which depends on intensity. So, intensity is going to also contribute to the phase matching. So, for partially ionized gas medium confined inside a waveguide, additional dispersion term due to waveguide is also in included and the magnitude of the propagation vector can be written as we have to include this term as well plus mu m n square lambda divided by 4 pi a square. (look the slides for mathematical expressions)

The in general, HHG process can be HHG frequency conversion can be obtained in a gas medium in a very short gas medium, in a very long gas medium or in an waveguide also.

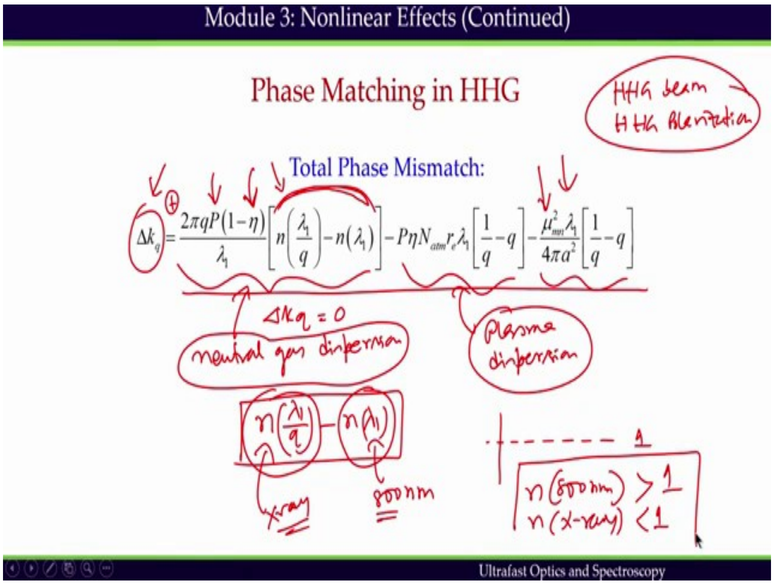
(Refer Slide Time: 15:31)



So, waveguide has a break here so we are in injecting gas molecules through this waveguide. So, if we have waveguide then we have to include the waveguide dispersion term as well in the medium. So, that goes in, if we do not use waveguide then we do not need this dispersion contribution we can use we can avoid that.

So, here what does it mean by this waveguide contribution a here is the inner radius of the waveguide and mu m n is the characteristics of the waveguide mode. For a gaseous medium this equation is sufficient; however, we have to consider waveguide contribution as well if we produce HHG in the in the waveguide.

(Refer Slide Time: 16:29)



So, the phase mismatch which we can write like this way by this expression and this expression gives us the phase mismatch between the emitted beam HHG beam HHG beam and HHG polarization that is that is going to control the phase matching. The phase mismatch can be given by this which has this expression here waveguide contribution we have included, if we are not using waveguide then we can we can we can remove this contribution.

Now, the phase matching of HHG can be achieved that is, we can have a condition delta kq equals 0 by balancing various parameters given in this equation, in this equation there are parameters which are looks positive, looks negative and looks negative. So, we can balance them positive negative such a way that in the end this delta k can be 0 and the parameters which you can control in this in this phase matching process is going to be pressure of course, pressure can be varied. Then ionization fraction, how many molecules we can ionize that can be varied and in the waveguide, we can also vary the mode quality of the waveguide that also we can vary.

Let us now take a closer look at these factors which can contribute to the phase mismatch because the harmonic yield is severely affected by phase mismatch. The first term on the on the in this equation arises due to phase mismatch caused by neutral gas dispersion. So, this part is neutral gas dispersion and this one is plasma dispersion, these are the two important contributions in the HHG process.

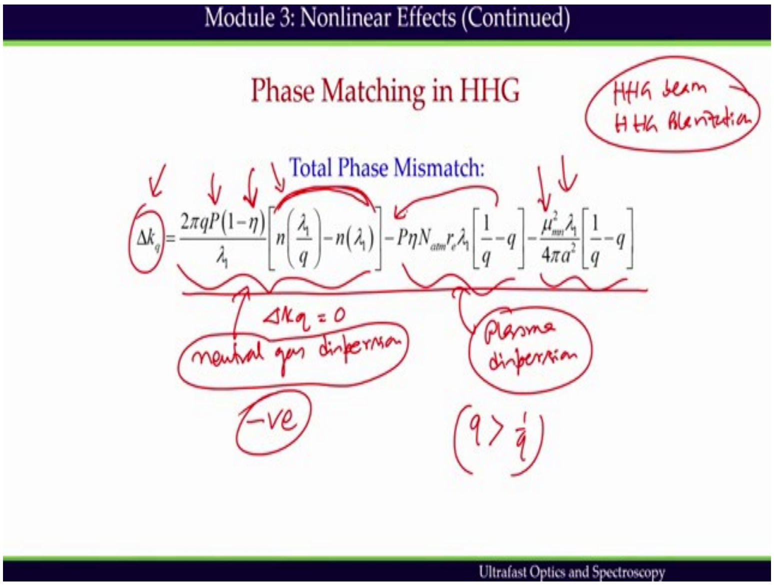
And we will see that by balancing these two distribution, we will be able to achieve this delta k equals 0. This neutral gas dispersion depends on pressure, difference of the refractive indices this on at the fundamental and harmonic wave length and ionization fraction. So, so there are three controlling factors I have for the neutral gas dispersion, P pressure 1 minus eta that is the amount of gas molecules would be ionized and the difference between these two at what wavelength of we are creating the HHG.

Ionization fraction which determines the ratio of neutral atoms and ionized atoms solely depends on the intensity of the fundamental laser beam. So, how much intensity we use that will control eta, but at the same time the neutral gas dispersion is negative. So, although it may appear to be positive, but it is actually a negative contribution, negative contribution is because the delta n what we have written here lambda 1 by q minus n lambda 1, this is negative always, why? Is because, the refractive index in the X-ray regime is less than 1. (look the slides for mathematical expressions)

So, this value is always less than 1 and this value is always greater than 1 this is the driving field. So, this refractive index is let us say 800 nanometre refractive index and this refractive index is going to be X-ray refractive index and in general the medium 800 nanometre refractive index is going to be always greater than 1 and sorry these are refractive index and X-ray refractive index is always going to be less than 1, that is an that is an import and there is an important consequences for this, for the X-ray optics we will not discuss those X-ray optics immediately.

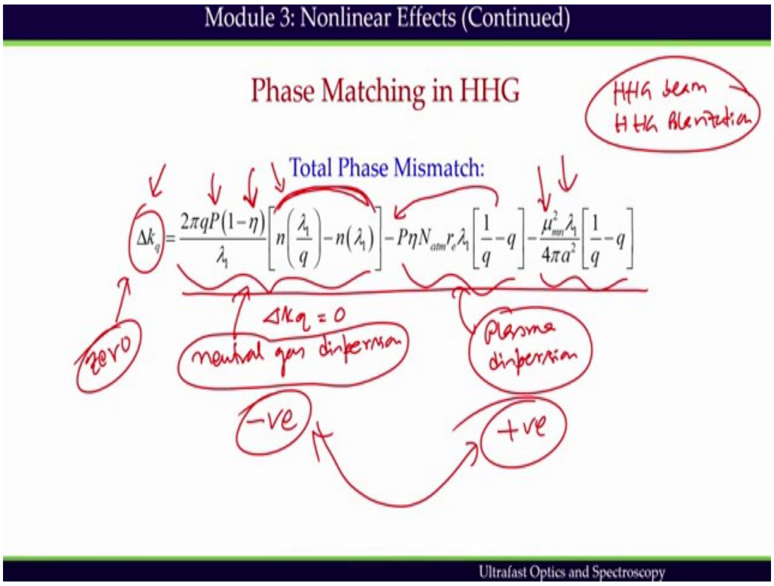
But what we see that, this term is actually negative term this refractive index difference in the neutral plasma dispersion and because it is negative the finally, neutral gas dispersion becomes negative.

(Refer Slide Time: 21:19)



So, this neutral gas dispersion instead of positive this, this term becomes negative. The second term which is plasma dispersion on the other hand is positive because q is greater than 1 by q. So, this is negative so this negative is contributing to this negative. So, finally, this is becoming a positive contribution.

(Refer Slide Time: 21:41)



When an intense laser beam interacts with the gas medium, free electrons are emitted. These free electrons cause plasma dispersion, as the contribution from the phase mismatch due to neutral gas dispersion is negative and due to plasma dispersion is positive, the total phase mismatch can be reduced by varying the laser intensity to adjust the ionization fraction. Considering the gas jet for the phase mismatch condition for phase match phase match matching condition, when the phase mismatch due to neutral gas dispersion is fully compensated by that due to plasma dispersion.

So, basically what happens? I want to make this to be zero and the only way I can make this to be zero is making balancing this positive and negative contribution to the phase mismatch and that will control the finally, the conditions which will fulfil the phase matching or the conversion efficiency in the HHG process.

With this we have come to the end of this module in this module we have discussed different non-linear processes, we started with linear versus non-linear polarizations, how they are affecting, how non-linear polarizations are creating new input beam and we have seen the phase matching is also an important concept in non-linear frequency conversion processes. So, if the phase matching is not achieved then we may not be able to get the efficient conversion for that particular conversion or non-linear conversion.

We have discussed polarization grating, we have discussed transient grating spectroscopy, we have discussed sum frequency generation, difference frequency generation and in the end we have discussed high harmonic generation which is a source which provides a source for ultrafast X-ray pulses.

With this we will end this talk and we will meet again for the next module.