Ultrafast Processes in Chemistry

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Module No # 08
Lecture No # 39
Brief Overview of Nonlinear Optical Phenomena (Contd.)

Let us complete a discussing that we have been performing over the last 2 modules we are discussing second order non-linear phenomena.

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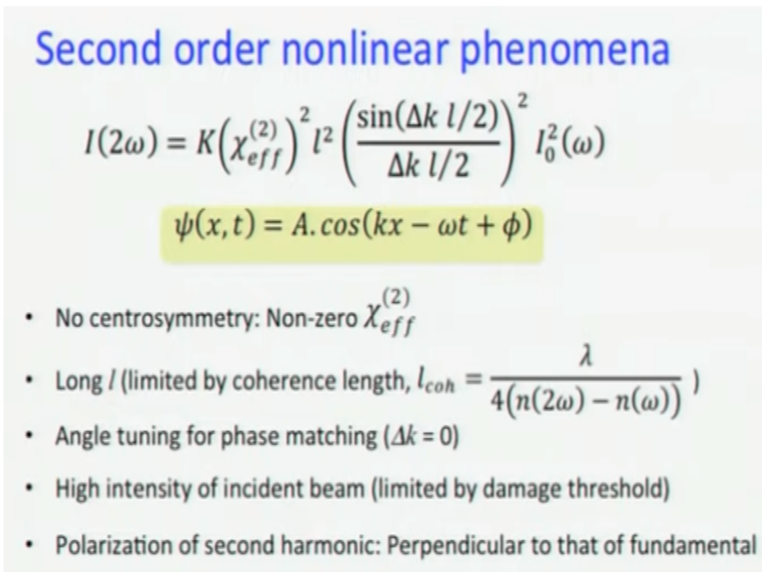

And we have said without deriving that intensity of the second harmonic light is proportional to square of second order non-linear susceptibility length of the crystal and something like sin square theta by theta square where theta involves delta k and I0 square of omega. And hence we have said that in order to get favorable conditions for getting second harmonic generation is you should use a high intensity of fundamental light.

You should use as much length of the second non-linear material as you can without messing up things you should have I mean high non I mean not you the medium should have a high second order non-linear susceptibility and finally delta k as to be equal to 0 that is what is going to continue up on delta k has to be equal to 0. What is k? Is a vector associated with the wave right k vector what is the meaning of delta k equal to 0?

Momentum is conserved in the process of second harmonic generation total momentum of the combining photons is equal to the momentum of the second harmonic photon okay. So from here the discussion we are performing is we want to know how it is possible to generate a second harmonic because one problem we had faced already is that the refractive index of 2 omega has to be equal to refractive index of omega and we have said in the earlier module that it is impossible to do it unless you have a birefringence crystal. So what we are going to learn in this module is how is it that birefringence helps?

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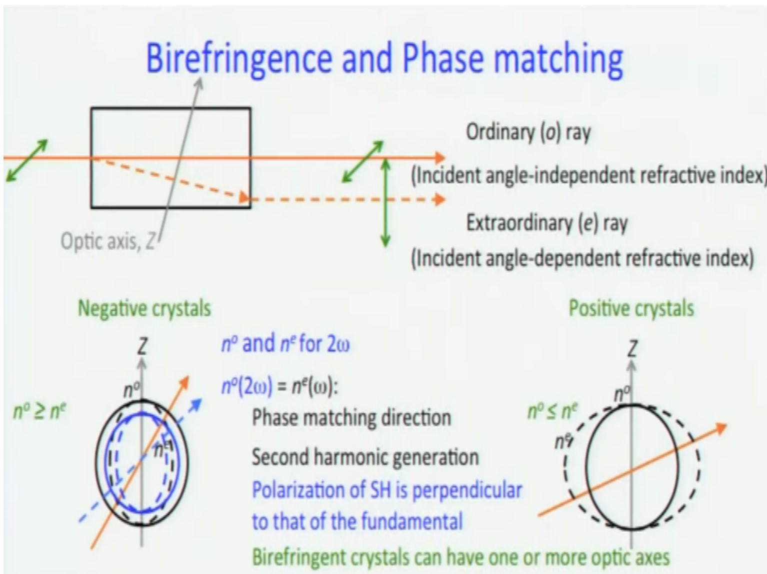

And we have already talked about birefringence but let us recap very quickly birefringence means this you have a crystal which is an optic access once again will tell you will discuss what optic access is? When light falls on it gets divided into 2 parts one of the rays that passes through is ordinary ray and the other one is extraordinary ray. In the ordinary ray the polarization is the same as the polarization of incident light in extraordinary ray polarization is perpendicular to that of incident light.

And then the most important thing is that for ordinary ray the refractive index of the material, please understand the refractive index of the material throughout is the same no matter what the angle of incidents is? However for the extraordinary ray refractive index on the angle of incidence so now what we will do is we will try to draw polar plots with only one circle or oval shape will not draw contours or anything.

Will try to draw a polar plots of this refractive index for o ray as well as e ray and we will do that for 2 different kinds of crystals as you will see. So let us say this is the optic access what is optic access? We will come to it wait a little bit longer and let us say this is the angle of incidence okay if optic access is Z and I call this angle between optic access and the incident ray theta does that remind you of something.

Optic access I am calling Z and I am calling the angle between the optic access and this incident ray to be theta does that ring a bell does that remind you or something? r theta Phi so I have already said it is theta right now position vector is done this is theta Azimuthal angle okay. That can be different right this is no special significance of making this theta small and making this theta large all we are trying to say is in principle theta can be anything from say 0 to 180 or even 0 to 360 in this case.

Now suppose for every value of theta a pens this is Z this is the incident ray this angle is theta so for every value of theta I am going to plot the refractive index once for the o ray once for the e ray once for the ordinary ray once for the extraordinary ray can you tell me what shape I will get for the ordinary ray. For ordinary ray if I plot so for this angle theta 1 I plot no and for this angle I plot no it will be the same is not it? Because remember for an ordinary ray refractive index is not dependent on the incident ray incident angle right.

So I will get a circle and it is a polar plot so do not so I will get a circle like this is the circle I get no where o stands for o ray ordinary ray. If I want to make a similar plot for the extraordinary ray what will it look like for an extraordinary ray remember n depends on the angle of incidence so ne if I call the refractive index of the extraordinary ray if I denote it as ne then it is going to have theta dependence it is not going to be circle what will it be?

Will it be a square? Will it be a triangle? What should it be? It will be a distorted circle so it will be an ellipse right. Because the point is this let us say this is the this is theta and let us say this is also theta this angle and this angle are same there is no reason why n should be any different for this position and this position right they will be same but the moment you change it will change in a different way.

So I am going to be have an ellipse but there are 2 cases that can be there and this is a good time to learn what an optic access is? An optic access is the direction in which if the incident ray propagates the direction of the optic access then refractive index of o ray and e ray are equal to each other okay. No separation between o ray and e ray if incidence is along the optic access that is what optic access is are you clear? Have you understood?

See no remains the same ne does not it changes but at some angle of incidence no and ne are equal that is the property that is there okay. So that line that you can get in that direction that defines the optic access right so that is your reference so what I am saying is with respect to optic access we are defining theta when theta = 0 then no = ne. When theta equal to 180 then also no = ne okay now with that I can draw 2 different ellipses see what I am saying is at this point and at this point ne and no have to be the same.

Or what happens in the middle ne can always be smaller than no or ne can always be larger than no. If ne is always smaller than no then I get an ellipse like this okay ne is shown in dashed lines and in the other case if at this moment I realize that and this point my animation could have been better the other case is for theta = 0 no and no = ne but after that ne is always more than no then this will be the case agreed in this one the first one no is greater than equal to ne.

The second one no is less than equal to ne when is no = ne in both the cases when is no = ne yes when the incident is along optic access or in other words theta = 0 or 180 degrees have you understood this diagram? Now let us get done with the definitions. In the first case when no is greater than equal to ne for those crystals we call them negative crystals. And of course if this is negative and then the other one crystals in which no is less than equal to ne those crystals are called positive crystals alright.

Now here let us say I have drawn this no and ne for the fundamental omega 1 now I want to draw no and ne for 2 omega but before going further are we all clear have you understood this part have you understood what is going on does it next introduce 1 phenomena already on this slide. Now after this we are going to introduce another, phenomena and they will add up to provide the condition for angle tuning for phase matching.

So it is important that we are all on the same page at this point is there any question is there any doubt everybody has understood everything can I go ahead? Okay now I want to draw the surfaces for no and ne for 2 omega okay and let us say I will draw it for the negative crystal you can draw it for the positive crystals yourself and it not very difficult also once we do it slowly. So first of all what will happen will a negative crystal become a positive crystal for the second harmonic or will it remain a negative crystal?

It will remain a negative crystal so we get similar kind of a diagram yes next question is well since no is a circle it is easier to talk about no is no going to be the same or different for 2 omega generally it is going to different we have already said that right. So is it is the circle going to be smaller or larger? For 2 omega is the refractive index larger or smaller that for omega it is smaller always smaller.

So what I will do now is I am going to draw that circle and ellipse for no and ne for 2 omega in the same picture is that I will draw it in blue okay understood let us go through this quickly once again have you understood this circle and ellipse business for no the polar plot is a circle because the refractive index does not depend on theta. Okay the polar plot means for this theta the tip of this pen will be at the value of n so if n is large it will be longer if n is small it will be like this okay.

So since no is the same for all values of theta the tip of this pen is going to define a circle however for ne it is not going to define a circle. With increase in theta it can either become smaller and smaller but when it becomes 90 degrees it goes beyond 90 degrees it will start becoming larger again until it gets the same value here okay. Or it become larger and larger until it reaches 90 degrees and then it becomes smaller again until at 180 degrees it is equal okay.

That is how we get this 2 pictures the first one the no is greater than equal to n1 why am I saying n1 and why am I saying no sorry where no refractive index for the ordinary ray is greater than equal to that for extraordinary ray those crystals are called negative crystals. And the second case where the refractive index for the ordinary ray is less than or equal to that for the extraordinary ray we call them positive crystals so far so good we understood this diagram.

Next we said no for the negative crystal you might said that we have drawn it for the positive crystal does not matter I just show you the example for the negative crystal now I want to draw this circle and ellipse for no and ne not for the fundamental omega but for the second harmonic 2 omega okay. So first thing we agreed upon is that this basic shape should not change a negative crystal will remain a negative crystal but then refractive index for 2 omega for the o ray is easier to understand o ray it will extend to e ray as well will be smaller than that for omega.

So if I draw the surface for no for 2 omega it is going to be a circle with a smaller radius and what about the surface for ne for the second harmonic it is going to a an ellipse but a smaller ellipse. So this is what I have drawn if possible try to not see the black picture try to see the blue picture only is the same as the black picture just smaller okay. Now comes the climax are you okay so far? Now see look very carefully there are 4 points at which no for 2 omega and ne for omega have the same value.

4 points at which no for 2 omega the blue circle as cut the surface for ne for omega the black dashed ellipse is that right? I will show you 1 look at this point and this is where it is important to understand polar plots what it means? The distance from the center that is the value and angular displacement from Z that is theta okay. So where the point where the blue circle and the black ellipse have over lapped is the point where their values are the same or in other words refractive index for the o ray of 2 omega is equal to refractive index of e ray of the fundamental okay.

That is a phase matching condition remember what was phase matching condition that the fundamental and the second harmonic have to have the same refractive index here it is achieved okay we set out with the problem that you take a regular crystal the refractive index for 2 omega will always be less than refractive index for omega. But then that problem has been circumvented because of birefringence.

Due to birefringence you produce 2 rays the ordinary ray and the extraordinary ray alright while the refractive index of the ordinary ray does not change for change in theta refractive index for extraordinary ray does otherwise It would have been just 2 circle that would have not overlapped. So because of this because you have 2 shapes you have an overlap some points of overlap 4 points of overlap to more precise 4 values of theta at which the refractive index for the ordinary ray with frequency to omega has the same value as the extraordinary ray for light with frequency omega.

So phase matching condition is achieved aright experimentally how will I do it what am I doing here? What I have discussed is this is Z optic access and this is the angle of incident we are talking about increasing heta this way no need I can move the optic access how will I move the optic access I will rotate the crystal. For the same angle of incidence let us say this is optic well we are using this for the optic access this is the incident beam and this is the optic access of the crystal I rotate until the appropriate theta value is achieved where you have no 2 omega = ne omega you are done okay.

This is why we get second harmonic generation conveniently by simply rotating the crystal what we are doing essentially is we are changing the angle between the optic access and the incident ray. Since Mohammed could not go to the mountain-mountain has come to Mohammed cannot change incident ray without messing up everything so we have just rotated the crystal. Now remember we have said something else we have said that the polarization of the second harmonic light is perpendicular to that of the incident light do you see why?

See phase matching condition has been achieved between what and what omega 2 omega I understand but ordinary ray of omega and sorry extraordinary ray of omega and ordinary ray of 2 omega and as we have said already o ray and e ray have perpendicular polarization. This is why second harmonic light has perpendicular polarization with respect to the fundamental okay. Polarization of second harmonic light is perpendicular to that of the fundamental okay.

So far so good everything is looking nice concluding comment for today is this now a days at least for commercial setup you only have everybody uses crystals of the single optic access uniaxial crystals there are crystals which are multi-axial fortunately people do not want to use them any more I have used and believe me it is a pain multi-axial means you have to turn not only along this but also along this and along this sometimes.

So if that happens you have more parameters and the problem is you never know you turn in 1 direction you do not know whether you have reached the correct direction or not because the other 2 directions are the other direction that is there maybe way off okay. If you can get everything correct then you will get very good second harmonic generation. But generally now a days in commercial systems people do not like to use anything other than uniaxial crystals.

For the simple reason that there is only one control you turn in 1 direction change this value of theta you can imagine if there is another axis what will happen there will be theta 1 and theta 2 and this theta is different from Azimuthal angle that in principle it goes from 0 to 360 I have shown 4 points right 4 angles at which you can get second harmonic generation do you observe that experimentally we do not right.

Usually get only at 1 sometimes you will get at 2 why? Because what it means is you want to go to the other side incident angle instead of going this way the light has to go this way so it is equivalent to turning the crystal completely who will do that nobody does it. So generally that is 1 unique point where you can get second harmonic generation so that is the end of this 3 module discussion what we hope to have achieved in this is that we have got ourselves introduced to the fascinating world of non-linear optics little bit without deriving anything.

And secondly we have at least understood 1 operation that we do regularly in lab and that is happily turn the crystal and see that you get nice blue light coming out. Now what we will do next is second harmonic is done in the next module will try to discuss a little bit about sum frequency generation and we will talk about 2 kinds of phase matching. And then we will go on to discuss the next step of second harmonic generation or sum frequency generation that is optical parametric amplification or optical parametric generation to start with.

See we are talking about generating sum frequencies so far it is also possible to generate different frequencies okay that is where we will get to and then when we are done with the discussion will try to discuss what is there inside the optical parametric amplifier and with that I think our discussion of instruments will be more or less done then we start discussing actual experiments we have already introduced pump probe but then what we can do is we can start with the classic experiments of Ahmed Zewail then go on to other pump probe experiments.

Then later on we are going to talk about experiments that involve to this non-linear properties as well you want to talk about Raman experiments and all. So we will need this later on as well.