Optical Spectroscopy and Microscopy
Prof. Balaji Jayaprakash
Centre for Neuroscience
Indian Institute of Science - Bangalore

Lecture – 41
Fundamentals of Optical Measurements and Instrumentation

Hello and welcome to the course on optical spectroscopy and microscopy, in this part of the lectures what we will be looking at is that way of generating new colours of light using nonlinear optics and this is one aspect of light generation that we would be utilising in order to generate the pump laser, pump light for the femtosecond system that we have seen that the using in our classes right, the titanium sapphire laser.

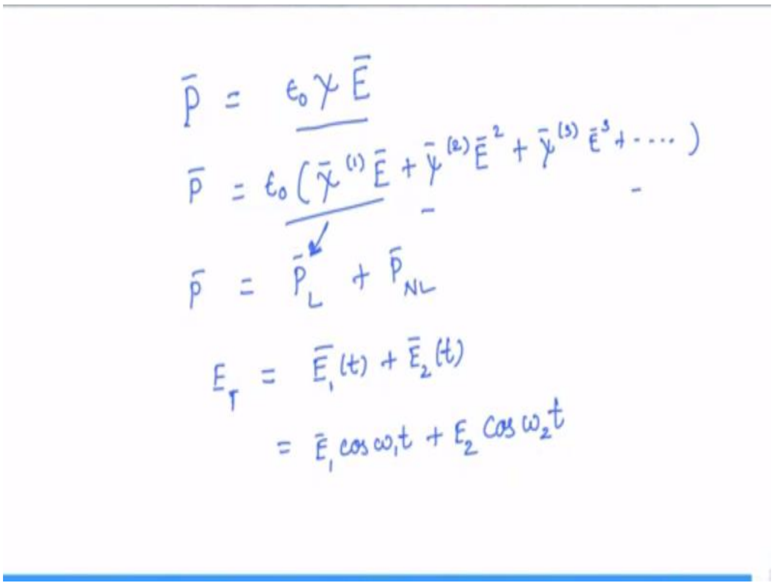
Titanium sapphire laser requires visible light to be pumped to the excited state, now there are different ways of achieving that I mean, one is to use an organ, a powerful organ in laser to pump that but modern day systems use a diode pumped solid sets lasers, they; what they do is they generate a light in the infrared region which goes and then pumps another laser 1064 laser and which is then used to generate the visible light.

Now, how is this visible light generation we will be looking at specifically but in that process to understand this process, we would develop a little bit of background on nonlinear optics and then new frequency generation to be more general right, okay. So, we know I mean the way we have talked about so far in the course have been in with respect to the light matter interaction is with respect to a molecule and how it is actually interacting with the electric, I mean the light itself.

Now, what we here going to do is that a microscopic body alright, just like a laser media or any substance now, if not entire media if you are going to take it and then say that okay, look now I am going to put this or place this whole media, the macroscopic system in an electric field and now what kind of a character that would have much, right that is the question we are going to ask.

In order to understand that the first thing is what you want to do is that we want to describe something called as a polarization vector, in the polarization that the molecules undergo or the microscopic substance that has developed because we have placed it in the electric field.

(Refer Slide Time: 03:19)



Now that polarization P can be written as in general, we know that the polarization P it is a vector dielectric polarization, it is a vector of the microscopic body can be written as epsilon 0 the permittivity of free space times the susceptibility of the molecule right, times the electric field in general, this description is pretty good when the electric field is not of higher magnitude nodes.

But when you start increasing the electric field such as you are having a very high intensity laser or you actually place the material inside a very high electric field like that of a pockel cell and so on and so forth, then it is; it turns out that this expression what we have written here is just an approximation to a much more broader and a much more general way of writing it, wherein we are going to expand this chi.

Because the chi itself start becoming dependent on electric field, so that we will start expanding the chi, you can expand the susceptibility and hence the entire polarization itself can be written down as the P epsilon 0 times chi 2 E square chi 3 E cube etc. So, now in such a case when you; in such a case, then this our first expression can be thought of as an approximation to the real situation, right.

Because when the electric field is; I mean usually the coefficients chi 2, chi 3 are small that when there is a moderate electric field, you do not see the effect of that in the polarization however, now the electric field magnitude has gone up to a considerable extent where even that small nonzero values of the chi start to contribute at a state here, it is not mandatory that all the materials have all of these values; the chi 2, chi 3 and etc., it is a tensor, in general it is a tensor.

And specifically, the centro symmetric molecules right, the molecules that processes an inversion symmetry we can actually, it goes back to our original descriptions of the light matter interaction writing down the matrix elements and then from the (()) (07:12), where we have seen we can by using the symmetry arguments we could set some of them as 0 and hence it does not happen and so on.

So, in that same light we can set chi 2 or in general or all the even chi's to be 0 for centro symmetric molecule or the molecules which have inversion symmetry alright, so for every molecule I mean the bottom line is every molecule can have different values of chi's and it is not mandatory that everybody should have a nonzero chi and in fact, there are class of molecules with do not have chi 2, I mean which have the chi 2's and all chi evens to be 0.

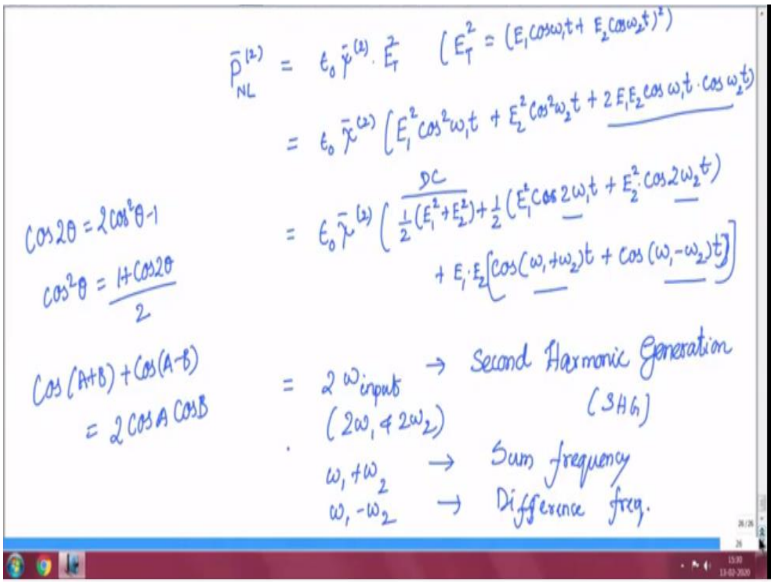
So, if you take a molecule and for this current discussion we will restrict ourselves to the chi 2 we just can think of that as a second order, so if we restrict our discussion to the second order and then ask what happens and what kind of a property does this chi 2 that this susceptibility bring in to this polarization itself and then how does it manifest in terms of light matter interaction that you will see soon that it possesses some very, very interesting properties.

And that is one of those properties that we will be utilizing to generate this new colours, so now let us concentrate ourselves on those substances that have nonzero chi 2 which are; which essentially by means that they are not centro symmetric, they do not possess an inversion symmetry, okay. So, in that in such a case we can write this P again as P; sum of P linear term plus P nonlinear where linear is basically this term while the nonlinear I am approximating everything outside; everything other than the first term.

First term basically, it is a scattering term but let us not worry about that so, we can collect all of the rest; I mean rest of the higher order terms as P nonlinear, it is a polarization; non-linear polarization induced because of the high because of the applied electric field. So, if you think of like this and then the, it happen that the electric field that has been produced is an oscillatory electric field just like 2 electromagnetic radiations of frequencies omega 1 and omega 2.

Then, we could take 2 of this E1 equals; the first E total equals the electric field as a function of time due to the first plus electric field as a function of time to the second one and we know for a plane wave, it is even cos omega 1t + E2 cos omega 2t. Now, using this 2 waves, 2 and then are; if the E the intensity is sufficiently high such as the electric field or E1 and E2 crossed by their intensity is able to induce a nonlinear polarization.

(Refer Slide Time: 11:42)



Then, we can write down that polarization and we will restrict to just chi2, the second term and not anything more, so to indicate that we can call that as polarization to the second order basically, it is a nonlinear polarization right, this corresponds to the nonlinear polarization but to the second order. If you do that, then we can write as epsilon chi 2, this need to be non 0 and E electric fields square, right.

And let us say, we are going to estimate this polarization inside the crystal because of this, this will be the total, so we can write this as epsilon 0 chi 2 times E1 square cos square omega 1t plus; by the way this instead of; so the field; electric field vectors and what we are actually when you are doing this thing what I am actually doing is I am writing it is the modulus; treating that as just a magnitude square.

So that is why I am, we can do that cos square omega 2t + 2 E1 E2, so cos omega1t times cos omega 2t, so what we have done is we have used this expression; expression 1 in here, so that we have the ET equals the from the first expression, so basically E1 square is; ET square is E1, sorry, I just use, omega t square, so using that idiom we can write it in this format which again can be simplified into or rewritten as; so we have mean, we have cos square omega 1t and cos square omega 2t.

And then there is this cos omega 1t + times cos omega 2t, so we can recognize here quite a few things which is cos of 2 theta is; sorry, we can write cos square theta equals 1 by 2 times okay, well actually let us write it from cos of 2 theta equals 2 cos square theta minus 1, so we have the cos square terms I would like to write it in terms of 2 theta, the idea is that we will see that there are new frequencies that frequencies in multiples of the original ones that are getting generated.

So, to recognize that we write we make use of the fact that it is cos square theta is 1 + cos 2 theta by 2, so and similarly cos of A + B + cos of A - B would be 2 times cos A cos B, so we will utilise, we will use these expressions to rewrite the cos omega terms as E1 cos square omega 1t can be written as 1/2 E1 square similarly, there will also be a E2 square coming from the second term and then we will collect that as a common.

And of course we can also collect 1/2 of E1 square cos 2 omega 1t + E2 square cos 2 of omega 2t + this term, the even this term now we can write it as cos of; of course (()) (18:14) E1, E2 term, so E1 dot E2 cos of omega 1 + omega 2t + we can write it as cos of omega 1 - omega 2t, thus you can actually see this, so the resulting polarization in principle contains or it looks like a very simple DC term, DC electric field that is this.

And then an AC components but the interesting thing is the AC components have different frequencies than what it has started with, 1 omega 1, 2 omega 2, omega 1 + omega 2 and omega 1 - omega 2, so we have 2 omega 1 and 2 omega 2's are in general 2 times the omega input is called a second harmonic generation and so this is this corresponds to 2 omega 1 as well as 2 omega 2, alright.

So, you could in principle think of these 2 components that we have started right, the E1 omega that the 2 plane waves E1 cos omega 1t + E2 cos omega 2t right, so these 2 plane waves I mean for the description here we have taken it to be at 2 different frequencies you could take a same one wave, omega 1; light of omega 1 frequency and then sufficiently high enough intensity where the second order term chi 2 starts to contribute you could think of that.

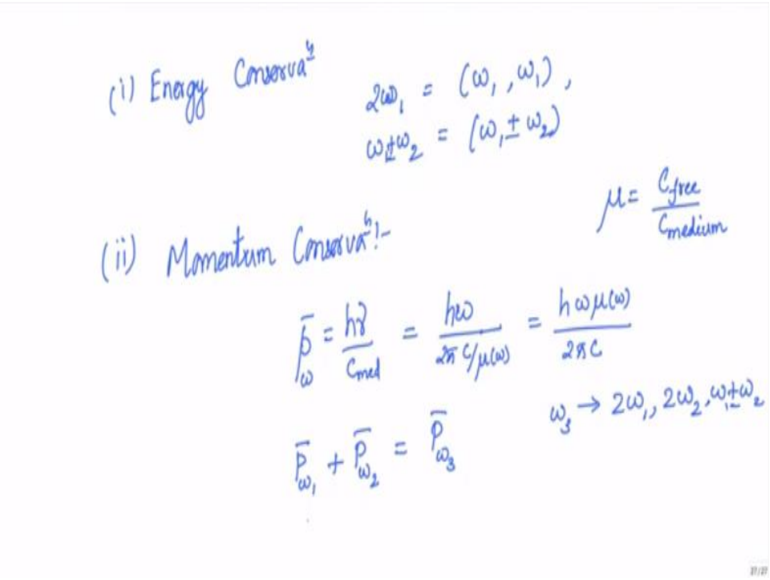
In that case, in such a case then what we have is all we have is just 2 omega 1 right, this is so you can see 2 omega 1, 2 omega 1, this will be 2 omega 1 and this will be 0 right, so that will be just the DC field again. So, now and since the intensity the electric field all will be equal that will just contribute to the DC itself but so this new colour, new harmonic is the second harmonic light or second harmonic generation, it is also called as SHG.

So but in general, it is good to write it as 2 omega the input, you can you also generate omega 1 + omega 2 which is the sum frequency omega 1 - omega 2 which is the difference frequency, so we started with 2 distinct colours of light, now what we have is that when we pass through a material which can have a nonzero chi 2 and then a sufficient; if you have sufficient intensity, what of these 2 components what we ended up generating are the colours of these new different frequencies; 2 omega 1, 2 omega 2, omega 1 + omega 2.

And omega 1 - omega 2, so you have multiple colours that gets generated, however one important aspect that we need to pay attention to is that the conservation laws her, so while in terms of energy conservation we have because it is ultimately I mean, even though we have for convenience sake we took the light as E omega 1 cos omega 1t, we know that we have established the equivalence of that description to that of the photons.

So, ultimately when you are talking about this new light generation you are generating those different frequency photons and in this scenario we need to make sure 2 conservations are met, conservation equations are satisfied.

(Refer Slide Time: 23:56)



One is energy conservation right, now that is fine because the energy, so 2 photons of omega 1, I mean 2 omega 1 is generated by utilizing photons of I mean, 2 photons of omega 1 and omega 1 and similarly, we can write down omega 2 is 2 omega 1, omega 2 and then the sum frequency omega 1 + omega 2 again the energy would be are plus minus omega 1 omega 2 would be utilizing 2 of this.

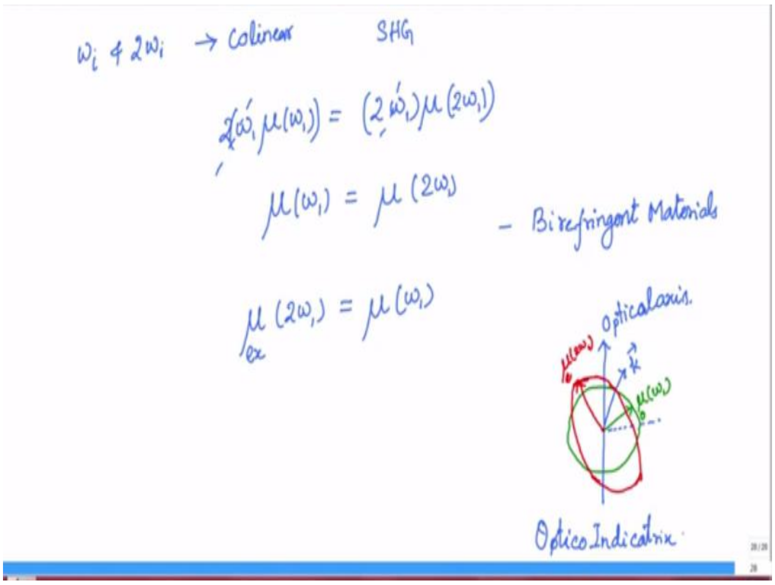
So, the new high energy photon, the higher energy comes from absorbing or annihilating 2 photons of lower energy whose sum gives rise to this new one, so that way the energy conservation is met however, it is not guaranteed that the momentum conservation is also met. Now, how do we go about checking this so, let us look at the momentum conservation, so the momentum of the photon we know is given by h nu divided by C at least and we can write this as h omega by 2 pi.

The velocity here is really the velocity in the medium, which means we have to; I mean the velocity in free space but what we have to do is we have to write it in terms of the medium. So, when we do this, so what we have is h omega, we have been using Mu, so let us stick to Mu, so h omega Mu function of omega divided by 2 pi C, I just used the definition of the refractive index here; refractive index Mu is basically in free space divided by C in medium.

So, C in medium is basically, C by Mu so, if you replace that we know the momentum of the photon in that medium, so traveling in the medium, so let us mark it here, so that is the momentum of a photon with frequency omega. So, in our case we have 3 different photons alright, 2 input photons so, we have P of omega 1 corresponding to the first P of omega 2 corresponding to the second and P the output of omega 3.

Now, this omega 3 could be any of this which could be 2 omega 1, 2 omega 2, omega 1 + omega 2 or omega 1 - omega 2 right, omega 1 difference omega 2 okay. So, now so for, I mean for a light we generated this condition had to be met now, what it means is that let us take the case of a second harmonic generation, so just omega 1 equals, I mean omega 3 equal to 2 omega 1.

(Refer Slide Time: 28:47)



And we also will make sure that the all of them right, that the photons of the omega 1 and 2 omega 1, omega input right, 2 omega i, both of them are collinear, let us make sure that they are traveling in the same direction, so this is very important right, so which means when we say this what we are talking about is that, collinear, so what you are saying is that the light that is generated is traveling along the same direction that is a the forward direction photons, right that is what we are looking at.

And then the expression becomes pretty simple, so what we are demanding is that 2 omega 1 Mu omega 1 equals 2 times the omega 1 Mu 1 right that is the idea here equals 2 omega 1 which is a new frequency here Mu of 2 omega 1, second harmonic generation that is collinear right, so that is what we are collinear second harmonic generation for such a condition we are looking for how can we match the; what it means to match the momentum, alright.

So, when we do this, what it implies is that at is; for we are looking for a special material such that the Mu of omega1 for this to be met right, you can actually see this that goes off, the Mu of omega 1 need to be equal to mu of 2 times this frequency. Now, the refractive index is a function of wavelength and the material right.

Now, what we are actually looking for is a special material whose refractive index either does not change with respect to the wavelength or it changes in its own way but somehow it has 2 refractive indices meaning it has 2 profiles; one is just a Mu of omega 1 and at some other profile you have this refractive index that somehow matches I mean, that is somehow numerically that is equal to that of the first one.

So, now when we talk about such kind of refractive index materials one I mean, it is basically we are talking about 2 different refractive index material; refractive index inside that material that is I mean you are talking about essentially, refractive index of the material for 2 different colours being the same, we know that we have come across materials called as birefringent materials, right.

So, these are materials whose which has refractive indexes, one that are different for ordinary and extraordinary polarizations right, or ordinary and extraordinary waves, so you could think of that the Mu of extraordinary wave for 2 omega 1 could be equivalent to Mu of omega 1, now but of course the extraordinary the refractive index depends on the propagation direction alright, while the regular Mu is independent of the propagation direction.

So, we could think we could represent this in a diagram wherein if you look at the, I mean it is basically orientation of the K vector right that is the direction along which the photons are being is coming out and going in and coming out, so we could represent this in a nice diagram where in use let us take a; let us mark the optical axis along the this vertical direction and then since the ordinary refractive index for ordinary wave is independent of the direction.

So, you can actually think of the refractive index would be same across any angle about any angle theta that we measure okay, this is the optical axis of the crystal now, let us say the light were to travel at K vector like that. So, now we since it is an independent along this direction that in this plane, so any direction that you pick and then rotate the crystal you would; the light would feel the same refractive index.

So, now this Mu that is maybe I should draw it in a different colour, so let us do that and I am going to draw it with a green, so I am actually drawing a circle because the refractive index is independent and it is symmetric right, so this represents the Mu 0 of omega 1, now I could in move this along any of this direction, the K along this entire rotate in this cone, it will have the similar refractive index for the ordinary wave.

However, the extraordinary wave has an orientation dependence and it is an ellipsoid, right that is what we know because it is a; so if you were to do that then so, let us say that okay, different colour here again, so the extraordinary wave is somewhere here and we could think of alright, so that being the extraordinary wave, which is Mu E of 2 omega 1, so while, I will draw it properly (()) (37:49), something like that.

And now, clearly depending on the orientation of this K vector you have a different refractive index of the different Mu for the extraordinary wave, so this Mu is E 2 omega 1, so by rotating the crystal alright, so you could reach a point where the refractive index of the extraordinary wave is equal to that of the refractive index of the ordinary wave and if you were to take a kind of a represent it in a top view manner, so you can think of this by the way, it is the overlapping, mean the points where this or the direction where this 2 touches or crosses each other.

And in such a condition what we have is that the omega 1 develops the polarization at in this material and all of these particles, the polarization are in phase such that, they generated 2 omega 1 adds up and gives you a light that is of frequency 2 omega 1 similarly, we could construct this kind of a diagrams are called optical indicatrix diagrams we could construct this optical indicatrix diagrams for optimizing at which orientation you would facilitate 2 omega 1, 2 omega 2 or omega 1 + omega 2 or omega 1 - omega 2.

All we need to do is sit down and write the corresponding momentum conservation equations and then say okay at which how do we actually match the refractive index or essentially what we are here trying to do is when you are trying to match the refractive index of change the orientation, we are actually trying to match the phase of this light that we are generating, so it is also called as a phase matching by rotating this crystal.

So, with that I think, I mean this is one of the example of generation of the second harmonic generation; second harmonic light, you can also do a third harmonic or any of that so, the basic principle underlying principle is exactly the same but we will use specifically this principle of second harmonic generation in the lab.

Or at least we will see an equipment that uses this to generate a visible light that converts the 1064 nanometer light, a laser light that is coming out from (()) (41:44) into a 532 nanometer light visible light, green light which can be used for the exciting the titanium sapphire laser, alright. Thank you and I will see you in the next class.