NMR Spectroscopy for Chemists and Biologists
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Lecture 35
Product Operator Formalism Continued

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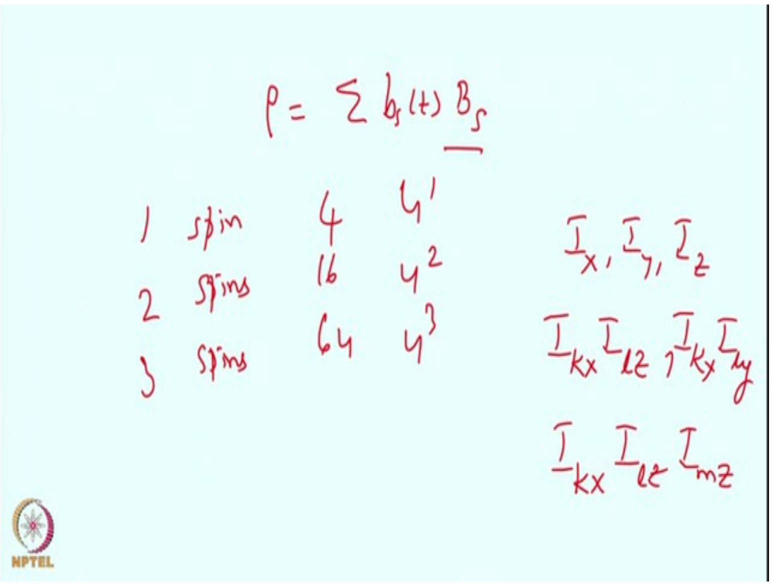

$$\rho = \sum b_s(t)\, \underline{B_s}$$

1	spin	4	4^1
2	spins	16	4^2
3	spins	64	4^3

I_x, I_y, I_z

$I_{kx} I_{lz}, I_{kx} I_{ly}$

$I_{kx} I_{lz} I_{mz}$

So, we have been discussing about the product operator formalism of the density operator which you wrote like this $\rho = \sum b_s\{B_s\}$ then we looked at the various basis operators, these ones. For one spin, for one spin we had a set of basis operators there were 4, for two spins their basis operators number were 16, for three spins then we have a number is 64, this is 4 to the power whatever that is, so this $4^1, 4^2, 4^3$.

And we also had said that these are all products of angular momentum operators and we also saw what these individual operators represent in the density matrix, where do they represent, we have various kinds of single operators and we also have operators like this which are products of two angular momentum operators and we will also have products of 3 angular momentum operators, I_{lx}, I_{lz}, I_{mz} and so on so forth.

So, these are some illustrations and we saw exactly what these individual, some of these individual operators represent for the one spin, two spin cases. Basically, we looked at those which are representing in phase magnetization of single spin and also for two spins.

We looked at in phase magnetization in all the three cases of one spin, two spin, three spin cases. We also looked at the products of this type that I_{kx}, I_{ly}, how they represent a mixture of double quantum and zero quantum coherences and by combination of more than 1 of such operators, we can generate pure double quantum coherence or pure zero quantum coherence.

So and we these ones therefore, they are present in the density operator looking at the density operator and looking at these ones we can make an interpretation of what is the system, how the system is evolving, through the pulse sequence what kind of coherences are generated through the pulse sequence.

This is the physical meaning of those, calculation of the time revolution we will come to it very soon. So, continuing with this discussion, what sort of spectra we expect for the anti-phase magnetization in the three spin case that is what we are going to see.

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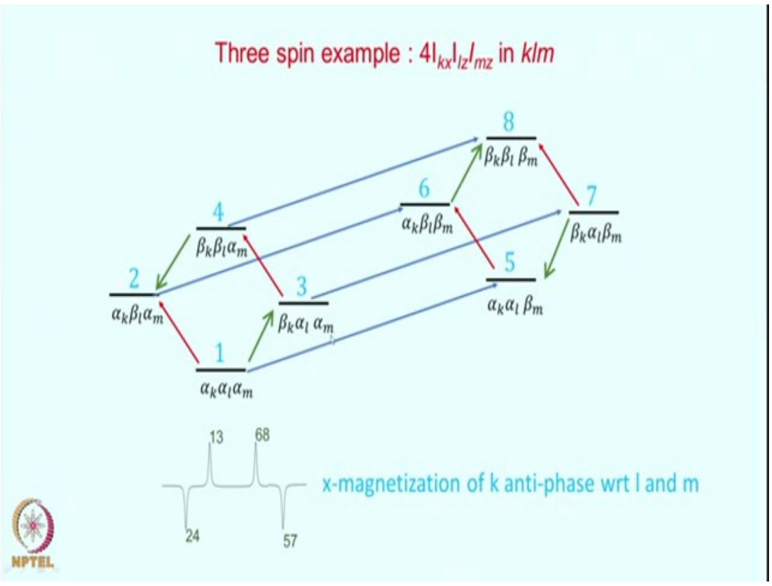

Three spin example : $4I_{kx}I_{lz}I_{mz}$ in *klm*

x-magnetization of k anti-phase wrt l and m

So, now we consider the three spin example. So, the three spin example we will represent it as *k l m* all the three are weakly coupled spins and we saw earlier in the three spin case also for operators like I_{kx}, I_{lx}, I_{mx}, what sort of transitions are occurring how do we represent this transitions in the three pin system. Let us do recap on the energy level diagram of the three spins case ones more. We have 8 energy levels here and these are represented as 1, 2, 3, 4, 5, 6, 7, 8 essentially it is a duplication of the two spin case.

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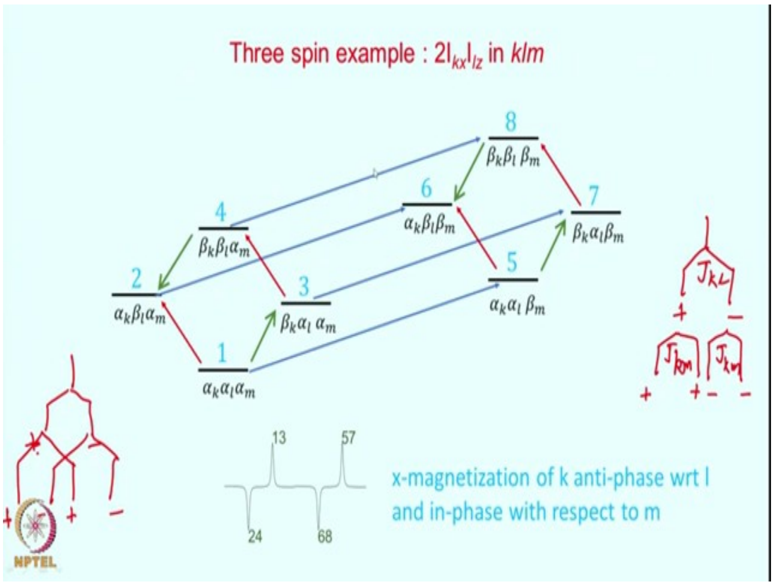
Three spin example : $2I_{kx}I_{lz}$ in *klm*

x-magnetization of k anti-phase wrt l and in-phase with respect to m

And in one of these four energy levels, you have the third spin, *m* spin as in the alpha state here and you see here this is all *m* spin is in the α state and here in these four energy levels, the *m* spin is in the β state, so that is how we generate the eight energy levels.

And the transitions accordingly are as *k* transitions and *l* transitions and m transitions. The ones which we connect from this group to this group are all the m transitions, the ones which are in this group they are basically the *k* and *l* transitions and that is indicated here. So, they are indicated by different colors here, so and therefore we can understand what these ones represents.

The arrows indicate what sort of spectra you will get when you actually measure the spectra of the three spin system and we have seen therefore in the earlier cases that we have in phase magnetization of the *k* spin if one is trying to see, then we will have four lines which are in phase and if we have similarly for the *l* spin and similarly for m spin.

So, now we are going to see, if I have an term of this type in the basis operator. For the three spin case, remember for the three spin case also we have 64 operators, 64 operators will consist of single angular momentum operators, the products of two angular momentum operators and products of three angular momentum operators.

Now, we are going to consider a two spin product for a three spin system and this is the operator like $2I_{kx}I_{lz}$ in the three spin system. What does it represent? So, it will be very similar to what we had in the two spin case itself that we will have these two transitions, there is one transition this is a *k* transition, α_k is going to β_k this is a *k* transition.

And correspondingly this is the *k* transition α_k to β_k but now you see the arrows, in this case the arrow is like this and in this case the arrow is like this. So, this represents an anti-phase term. These two terms will have opposite signs and that is indicated by gain of a representation here.

If 1 3 is positive then the 2 4 transition is negative and that will be in this block. The same thing will happen in this block as well 5 7 and 6 8 will also have opposite signs, if this is 5 7 is positive, 6 8 will be negative. So, therefore you have here 5 7 transition and 6 8 transition. The separation between these two will be the coupling constant between the spins *k* and. The same thing is here, of course the separation between these two or these two will be the coupling constant of the spin *k* with spin *m*.

So, you have therefore a doublet of a doublet and you can see here the anti-phase nature is for the coupling of *k* and *l*, whereas if you look at these two peaks which arise as result of the coupling between *k* and *m* they are in phase. So, therefore with respect to the *m* spin these are in-phase, these whole doublet is, has the same sign as this doublet.

This is negative or positive and this is negative. The same sign pattern here for this one and this one. So, therefore we called this as the *k* magnetization which is anti-phase with respect to *l* but in-phase with respect *m*. Let me repeat that again here, so we have here four lines, the four lines come as a result of coupling between *k* to *l* and *k* to *m*.

And we can represent here the same in a slightly different notation, I can write that here itself, so let say I have one line for the *k* spin in the absence of any coupling, then I will have here two lines as a result of coupling J_{kl} and let me represent this as ± because I get because of the *k* in this system, we will have 1 3 transition and 2 4 transitions are have the opposite signs.

Now, there is a *km* coupling subsequently. So, *km* coupling results to splitting like this then we will have situation like this, then we will have this one we called it as [+,+] here and [- -] here and this is *km* coupling. So, in this case I have assumed that *kl* coupling is larger than *km* coupling.

But you can also have a situation where *km* coupling is larger than the *kl* coupling, so in that case what one could also do is we have this here then we have these two as the result of the *kl* coupling and then we will have the splitting again due to the km coupling, then we will have this sort of a situation.

So, these then will be [+ -], this will be [+ -] because this is [+ -], this splits into two when then it will give to the [- -] here and this + splits into two then we get [+ +] here. So, this is the kind of the situation what you are saying in this diagram.

So, this is what you are saying in this diagram, we have these two transitions are shown here as [+ -] that is this situation and this is [+ -] and + whatever that is you may call this [+ -] or vise versa does not matter.

So, you get different kinds of atoms depending upon the relative magnitudes of the coupling constants. So, here we have assumed the km coupling is a larger than the *kl* coupling. The *kl* coupling is resulting in this kind of anti-phase peaks which are positive and negative here and this is anti-phase peaks and this peaks are in-phase because this is due to the km coupling if this is positive positive or negative negative and this is negative negative or positive positive correspondingly.

So, therefore this is the way you get for the *kx lz* in a three spin system. If you were to take *ky lz* in the similar situation you will get the same sort of pattern in both this cases but they will have the dispersive line shape because of the *ky* term, if I take a *ky* term it will have a dispersive line shape. So, similarly for if I were to take *ly mz* or *my lz* so accordingly one will have such kind of patterns in the individual multiplets of the *l* and the *m* spins. So, this is an illustration to show how the different transitions appear in your spectrum.

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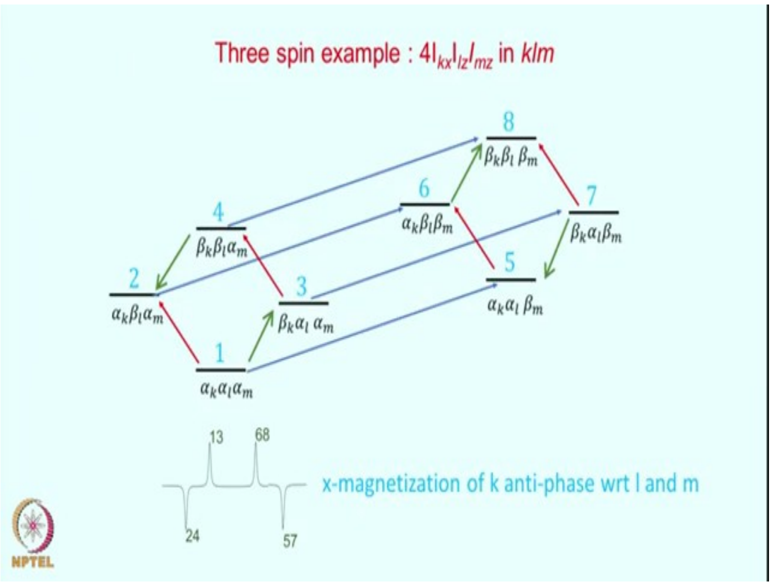
Three spin example : $4I_{kx}I_{lz}I_{mz}$ in *klm*

x-magnetization of k anti-phase wrt l and m

Now, let us consider a further extension of this, we will consider three spin product. A particular example I have taken here is the three spin product I_{kx} I_{mz} I_{lz}, 4 I_{kx}, I_{lz}, I_{mz}. Notice here, the transverse is only for the *k* whereas the *l* and the *m* spins are along the *Z* axis.

Therefore, this ones, this term obviously represents the *k* magnetization, because whatever is in the transverse plane is what we observe and we called that it represents that magnetization. So, this is the *k* magnetization. Now, we can intuitively we can think that this is something that is anti-phase with respect to both the *l* spin as well as the *m* spin.

So, what do you expect here, so in this situation earlier this group was had the same sign as this group, if this is negative positive this is also negative positive that is because we had in phase character with respect to the *m* spin.

Now, the *k* magnetization in this situation will be anti-phase with respect to m as well. Therefore, the first anti-phase term appears because of this, the *kl* splitting and secondly this whole term is inverted because of the anti-phase nature with respect to the *m* spin, therefore this now which was negative here, now becomes positive and this one becomes negative.

Therefore, you get a pattern here this is negative, positive, positive, negative. The coupling constant here is of course this is the *kl* coupling and from here to here that remains the km coupling. Now, we see, if you look at the splitting due to the *km* it is one line here another line here.

Now, these two are having opposite signs with respect to each other and therefore this is anti-phase with respect to the *km* splitting as well. So, it is anti-phase with respect to the *kl* splitting, this is negative positive *kl* splitting, it is also anti-phase with respect to the *km* splitting if this is negative and this is positive. Similarly, this and this have opposite sign, therefore we called this as anti-phase magnetization of *k* with respect to both *l* and the *m* spins.

So, in the diagram here you can also see, if I take this as a positive direction here this as a negative direction here and then correspondingly I will have this as negative direction here and this has a positive direction here. The same thing is represented schematically this is the schematic representation of how the peaks will appear in your spectrum when we record, when your density operator contains this sort of a term.

This sort of a term is a basis operator present in our density operator then if you were to measure this then you will get spectrum which will produce a spectrum like this. This particular term of course where as other terms will produce different kind of spectra. So, we are interpreting how, what are the contributions of the individual basis operators in your NMR spectrum.

So, therefore so much for the physical meanings of the individual basis operators, what do they represent in your NMR spectrum? But now we come to the next objective that is we must be able to calculate the evolution of the density operator consisting of all of this basis operators through the pulse sequence.

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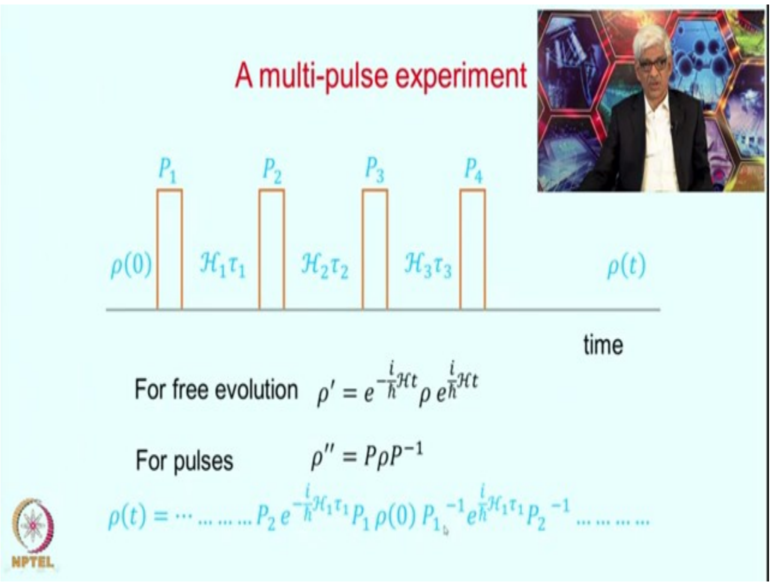

A multi-pulse experiment

For free evolution $\rho' = e^{-\frac{i}{\hbar}\mathcal{H}t} \rho \, e^{\frac{i}{\hbar}\mathcal{H}t}$

For pulses $\rho'' = P \rho P^{-1}$

$\rho(t) = \cdots \ldots \ldots \ldots P_2 \, e^{-\frac{i}{\hbar}\mathcal{H}_1\tau_1} P_1 \, \rho(0) \, P_1^{-1} e^{\frac{i}{\hbar}\mathcal{H}_1\tau_1} P_2^{-1} \ldots \ldots \ldots \ldots$

I again repeat this slide here, this slide you have already seen before, this is the multi-pulse experiment with so many pulses and the system is evolve with the time starts with the time 0, we have this pulse here and $H_1\tau_1$ tau1 pulse here then evolution under free precession for the Hamiltonian $H_2\tau_2$, then P_3 $H_3\tau_3$, P_4 and so on so forth.

So, for free evolution we have to calculate the evolution under the Hamiltonian this is for these ones and then for the pulses we have this evolution under the pulses $P_1 \rho P_1^{-1}$and ρhere can be a summation of all the basis operators and this is the same equation what we had earlier start with $\rho(0)$ keep applying the pulses and the evolution under the various Hamiltonians as we have discussed before.

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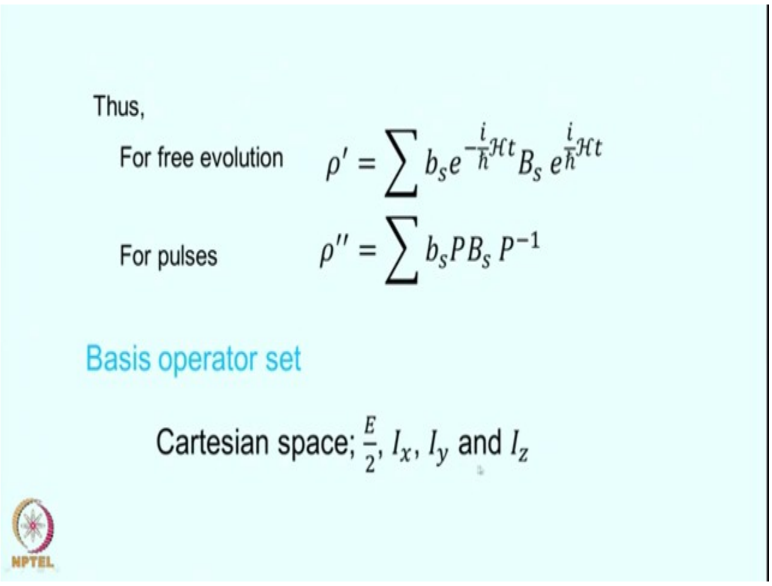

Thus,

For free evolution $\rho' = \sum b_s e^{-\frac{i}{\hbar}\mathcal{H}t} B_s\, e^{\frac{i}{\hbar}\mathcal{H}t}$

For pulses $\rho'' = \sum b_s P B_s\, P^{-1}$

Basis operator set

Cartesian space; $\frac{E}{2}$, I_x, I_y and I_z

So, now we explicitly put this basis operators here for the free evolution, I called this as

$$\rho' = \sum b_s e^{-\frac{i}{\hbar}\mathcal{H}t} B_s\, e^{\frac{i}{\hbar}\mathcal{H}t}$$

and for the pulses we called it as

$$\rho'' = \sum b_s P B_s\, P^{-1}$$

The basis operator set for the single spin, we have this expression when all the multiple spins are also constructed from this individual angular momentum operators as I mentioned before.

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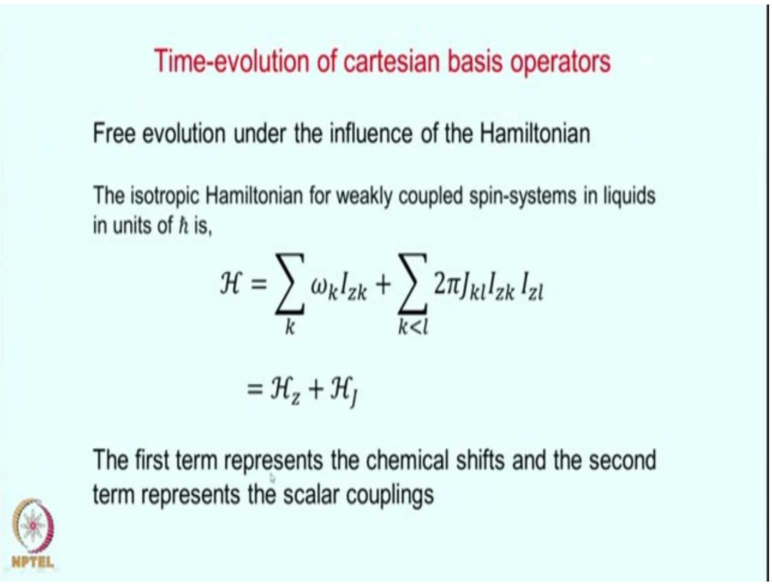

Time-evolution of cartesian basis operators

Free evolution under the influence of the Hamiltonian

The isotropic Hamiltonian for weakly coupled spin-systems in liquids in units of $\hbar$ is,

$$\mathcal{H} = \sum_{k} \omega_k I_{zk} + \sum_{k<l} 2\pi J_{kl} I_{zk} I_{zl}$$

$$= \mathcal{H}_z + \mathcal{H}_J$$

The first term represents the chemical shifts and the second term represents the scalar couplings

Now, let us go further and try and calculate the evolution under the Hamiltonian and evolution under the pulses. What happens when you apply the pulses, what happens when the density operator evolves under the Hamiltonians. Now, let us write the Hamiltonian ones more here explicitly we will write the isotropic Hamiltonian for weakly coupled spin systems in liquids consist of two terms and this is the Zeeman term where it is due to the interaction between the nuclear magnetic moments and the magnetic field and this is represented as

$$\mathcal{H} = \sum_{k} \omega_k I_{zk} + \sum_{k<l} 2\pi J_{kl} I_{zk} I_{zl}$$

the k is the index which goes over the various spins.

So, we can write as *kz* or *zk* does not matter, earlier we have used *kz* here but does not matter, it means the same thing we are referring to the same thing. This is the precessional frequency of the case spin and this is the coupling constant term, where we have J_{kl} is the coupling constant and the operator term for the interaction is $I_{zk} I_{zl}$ or you may call it as $I_{kz} I_{lz}$, so it is the same thing just indices are changed here.

So, we could have written as $I_{kz} I_{lz}$ as well same thing. And in briefly we write it as

$$= \mathcal{H}_z + \mathcal{H}_J$$

This is the complete Hamiltonian this is the Zeeman Hamiltonian, the first term represents the chemical shifts and the second term represents the scalar couplings.

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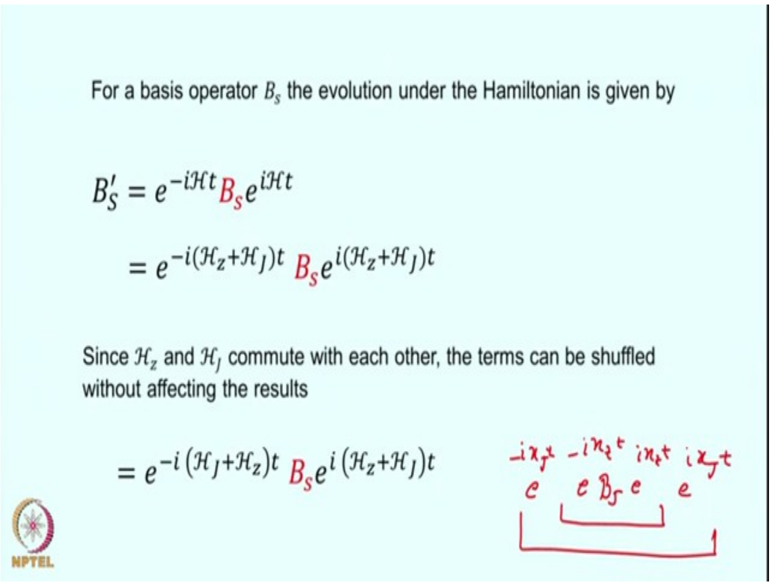


For a basis operator B_s the evolution under the Hamiltonian is given by

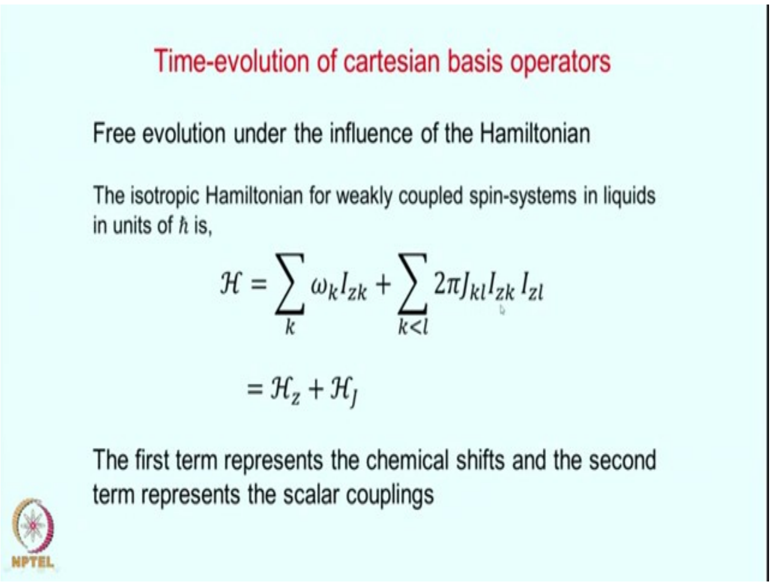
$$B_S' = e^{-i\mathcal{H}t} B_s e^{i\mathcal{H}t}$$

$$= e^{-i(\mathcal{H}_z+\mathcal{H}_J)t}\, B_s e^{i(\mathcal{H}_z+\mathcal{H}_J)t}$$

Since $\mathcal{H}_z$ and $\mathcal{H}_J$ commute with each other, the terms can be shuffled without affecting the results

$$= e^{-i(\mathcal{H}_J+\mathcal{H}_z)t}\, B_s e^{i(\mathcal{H}_z+\mathcal{H}_J)t}$$

$$\underbrace{e^{-i\mathcal{H}_J t}\,\underbrace{e^{-i\mathcal{H}_z t} B_s e^{i\mathcal{H}_z t}}_{}\, e^{i\mathcal{H}_J t}}_{}$$



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$$= \mathcal{H}_z + \mathcal{H}_J$$

The first term represents the chemical shifts and the second term represents the scalar couplings

For evolution of a basis operator Bs obviously, we can write this Bs prime this for the rho prime I write it as

$$B_S' = e^{-i\mathcal{H}t} B_s e^{i\mathcal{H}t}$$

This is the very simple extension of the previous equation.

For $\mathfrak{H}$ now I write explicitly

$$= e^{-i(\mathcal{H}_z+\mathcal{H}_J)t}\, B_s e^{i(\mathcal{H}_z+\mathcal{H}_J)t}$$

So, basically putting for $\mathfrak{H}$ here summation sum of the Zeeman and the coupling terms. Now, for the weakly couple spin system you remember what the Hamiltonian consisted of, so Hamiltonian consisted of I_{kz} term here and kz lz here.

So, these are all z components, the z components therefore this part of the Hamiltonian commutes with this Hamiltonian because kz operator, the z operators they all commute and they all belongs to the same spin. So, therefore these two will commute. So, when e do that then of course we can actually calculate the evolutions of this operators separately without affecting the result in any manner.

So, if I were to write explicitly this, I can write this $(H¿¿Z+H_J)t¿$ $(H¿¿J+H_Z)t¿$ does not matter because it only means there is an order of the order of the calculation is going to be interchanged. So, here I keep the same $(H¿¿Z+H_J)¿$, this is $(H¿¿Z+H_J)¿$and this part I turn it in the reverse manner, I write it as $(H¿¿Z+H_J)¿$.

What is the consequence of this? For the

$$= e^{-i\,(\mathcal{H}_J+\mathcal{H}_Z)t}\ B_S e^{i\,(\mathcal{H}_Z+\mathcal{H}_J)t}$$

The central term will represent chemical shift evolution.

So, let me write that explicitly here, so we write it as B_s e^{-iH_Zt} e^{iH_Zt} and then here it is e^{-iH_Jt} and e^{iH_Jt}. So, this particular term here, this is the chemical shift evolution and from here to here this after that this will be the J evolution, so we will first calculate what is the chemical shift evolution of a particular basis operator B_s.

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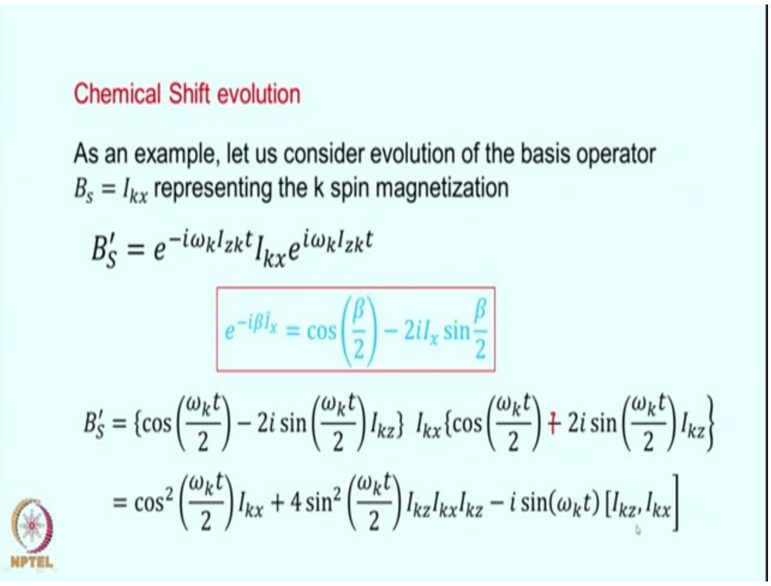

Chemical Shift evolution

As an example, let us consider evolution of the basis operator $B_s = I_{kx}$ representing the k spin magnetization

$$B'_S = e^{-i\omega_k I_{zk} t} I_{kx} e^{i\omega_k I_{zk} t}$$

$$\boxed{e^{-i\beta \hat{I}_x} = \cos\left(\frac{\beta}{2}\right) - 2iI_x \sin\frac{\beta}{2}}$$

$$B'_S = \{\cos\left(\frac{\omega_k t}{2}\right) - 2i\sin\left(\frac{\omega_k t}{2}\right) I_{kz}\}\ I_{kx}\{\cos\left(\frac{\omega_k t}{2}\right) + 2i\sin\left(\frac{\omega_k t}{2}\right) I_{kz}\}$$

$$= \cos^2\left(\frac{\omega_k t}{2}\right) I_{kx} + 4\sin^2\left(\frac{\omega_k t}{2}\right) I_{kz}I_{kx}I_{kz} - i\sin(\omega_k t)\,[I_{kz}, I_{kx}]$$

So, for this we take an example of Ikx. Let us say $B_s = I_{kx}$ and we will calculate the chemical shift evolution, that is under the influence of the Hamiltonian H_Z. So, the

$$B'_S = e^{-i\omega_k I_{zk} t} I_{kx} e^{i\omega_k I_{zk} t}$$

So, essentially we are meaning the same thing. So, this will be

$B'_s = e^{-i\omega_k I_{kz} t} I_{kx} e^{i\omega_k I_{kz} t}$

We have previously derived this formula,

$$\boxed{e^{-i\beta \hat{I}_x} = \cos\left(\frac{\beta}{2}\right) - 2iI_x \sin\frac{\beta}{2}}$$

this expression we have derived earlier by explicitly expanding this exponential function as a series and then we figured out that this can be represented as a simple manner like this.

So, we will use the same trick here. So, we will write explicitly this one in the form of this kind of an equation,

$$B'_S = \{\cos\left(\frac{\omega_k t}{2}\right) - 2i\sin\left(\frac{\omega_k t}{2}\right) I_{kz}\}\ I_{kx}\{\cos\left(\frac{\omega_k t}{2}\right) + 2i\sin\left(\frac{\omega_k t}{2}\right) I_{kz}\}$$

So, let us explicitly calculate this. We get here

$$= \cos^2\left(\frac{\omega_k t}{2}\right) I_{kx} + 4\sin^2\left(\frac{\omega_k t}{2}\right) I_{kz}I_{kx}I_{kz} - i\sin(\omega_k t)\,[I_{kz}, I_{kx}]$$

So, this is $-2i$ and $+2i$ gives me $-4i^2$ that is + 4 and since we get $\frac{t}{2}$ but this operators they can move around I have to keep them here, I_{kz} I_{kx} and I_{kz}. So, these ones will stay here, this do not commute and therefore I cannot move the around and then I will add here two terms of the cross term one due to this and the other one due to this, this gives me $-\sin\omega_k t$ a commutator of I_{kz}, I_{kx} the two terms club together can be represented in this manner so I have a commutator here I_{kz} I_{kx}.

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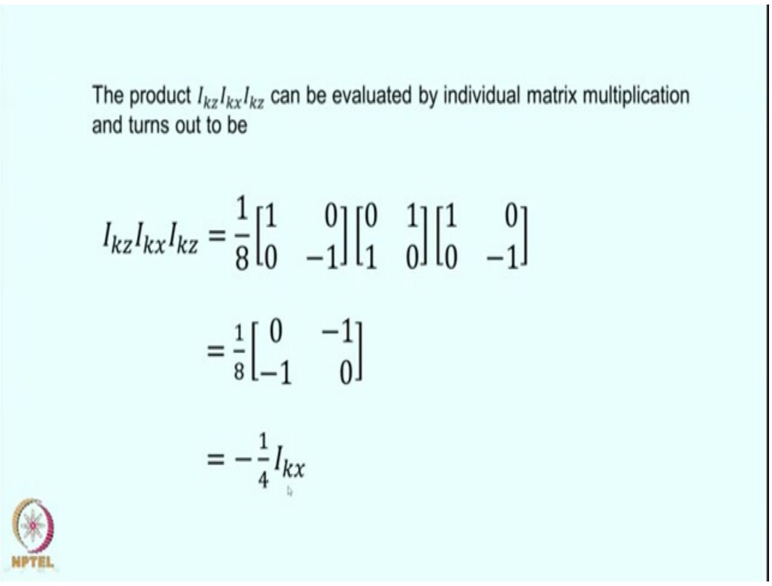

The product $I_{kz}I_{kx}I_{kz}$ can be evaluated by individual matrix multiplication and turns out to be

$$I_{kz}I_{kx}I_{kz} = \frac{1}{8}\begin{bmatrix}1 & 0\\ 0 & -1\end{bmatrix}\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\begin{bmatrix}1 & 0\\ 0 & -1\end{bmatrix}$$

$$= \frac{1}{8}\begin{bmatrix}0 & -1\\ -1 & 0\end{bmatrix}$$

$$= -\frac{1}{4}I_{kx}$$

Now, here we have to calculate what this three product term implies, so we have here I_{kz}, I_{kx} I_{kz}. What does this imply? So, let us try and calculate that using this matrices once more, I have put here I_{kz} is

$$I_{kz} = \begin{bmatrix}1 & 0\\ 0 & -1\end{bmatrix}, I_{kx} = \begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}$$

So, you put this three matrices and because this is the k spin I can simply take the product. Notice all of these belongs to the *k* spin, therefore I can simply take the product here and this gives me $\frac{-1}{4}I_{kx}$.

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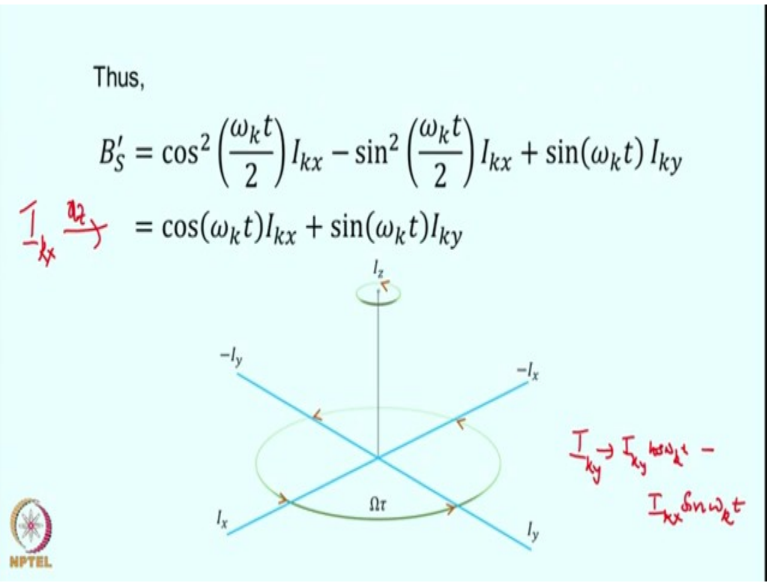
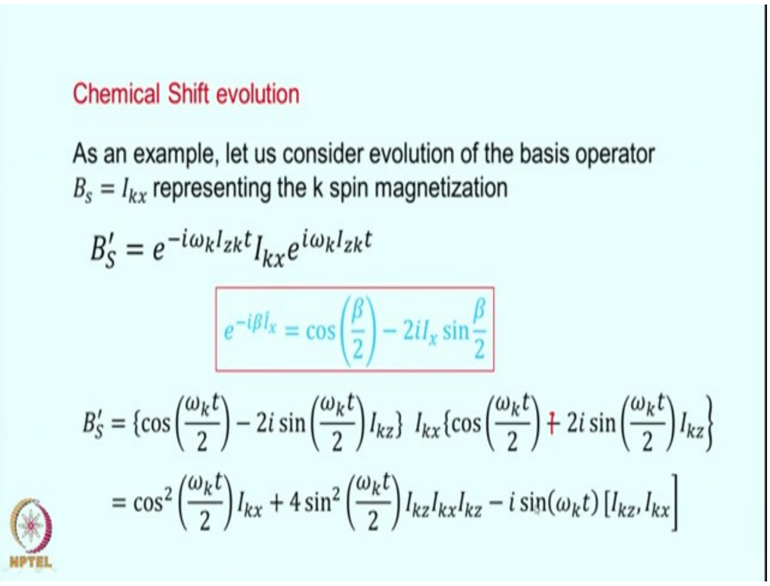


Thus,

$$B'_S = \cos^2\left(\frac{\omega_k t}{2}\right) I_{kx} - \sin^2\left(\frac{\omega_k t}{2}\right) I_{kx} + \sin(\omega_k t)\, I_{ky}$$

$$I_{kx} \xrightarrow{\Omega_z} \quad = \cos(\omega_k t) I_{kx} + \sin(\omega_k t) I_{ky}$$

$$I_{ky} \to I_{ky}\cos\omega_k t - I_{kx}\sin\omega_k t$$



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$$\boxed{e^{-i\beta I_x} = \cos\left(\frac{\beta}{2}\right) - 2iI_x \sin\frac{\beta}{2}}$$

$$B'_S = \left\{\cos\left(\frac{\omega_k t}{2}\right) - 2i\sin\left(\frac{\omega_k t}{2}\right) I_{kz}\right\} I_{kx}\left\{\cos\left(\frac{\omega_k t}{2}\right) + 2i\sin\left(\frac{\omega_k t}{2}\right) I_{kz}\right\}$$

$$= \cos^2\left(\frac{\omega_k t}{2}\right) I_{kx} + 4\sin^2\left(\frac{\omega_k t}{2}\right) I_{kz} I_{kx} I_{kz} - i\sin(\omega_k t)\left[I_{kz}, I_{kx}\right]$$

So, therefore that 4 4 cancels then I will get here for

$$= \cos^2\left(\frac{\omega_k t}{2}\right) I_{kx} + 4\sin^2\left(\frac{\omega_k t}{2}\right) I_{kz} I_{kx} I_{kz} - i\sin(\omega_k t)\left[I_{kz}, I_{kx}\right]$$

Therefore, what I got finally, I had

$$= \cos(\omega_k t) I_{kx} + \sin(\omega_k t) I_{ky}$$

So, after all this calculation what we have got is I_{kx} as a result of evolution under the I_{kz} Hamiltonian I get this two terms, say if I want to write here in simple manner an as in symbolic form I_{kx} under the influence of the Hamiltonian H_Z gives me it simply means the rotation gives me I_{kx}.

So, I write to represent this in the pictorial manner like this. Let us say we draw the three axes. The three axes we label them as I_x I_y and I_z. Of course it can be is a very general thing here, it can be *k* spin *l* spin whatever it is, so I just represent it as *x y* and *z* and this represents the Hamiltonian part.

The circle here mean implies this is the Hamiltonian and I have the transverse magnetization this is the basis operator here *kx* here the basis operator and under the influence of Hamiltonian this is the time evolution that is going on, this rotates in this manner. So, after a certain time t if your rotation is here, then I have a

$$= \cos(\omega_k t) I_{kx} + \sin(\omega_k t) I_{ky}$$

So, this represents a rotation in this manner, so you can extend this argument further to say that if I were to take I_{ky} here suppose it were I_{ky}, the same rules will apply so this represents the full rotation of the entire magnetization components.

So, if I were to start from I_{ky} here, then after the certain time tau if the magnetization is rotated like this then what I would get out $I_{ky} \cos \omega_k t$, I_{ky} because this will be cosine, if this it has rotated like this, so therefore I will have this component $\cos \omega_k t I_{ky} - I_{kx} \sin \omega_k t$ in that case.

So, if I were to write for the I_{ky}, let me also write that here, so if I were to start with I_{ky} then this will go $I_{ky} \cos \omega_k t - I_{kx} \sin \omega_k t$. So, that represents the rotation of the y magnetization from here to here, so therefore we have derived this principle to how this evolution can occur. So we stop here.