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PROFESSOR: So, good afternoon. Today we want to wrap up our discussion on two-photon processes. And just to repeat my motivation that in almost all cases when you address atoms, you do two photon courses because a photon is scattered. You may think it's absorbed and emitted, but in reality, it is a two photon process and not two single photon processes. So therefore, you should really pay attention. If you have any doubts about some subtleties about how is light absorbed and emitted, the correct answer is always obtained from the two-photon picture.

Now, I'm using pre-written slides because we treat two-photon absorption in perturbation theory. And it is exactly the same perturbation theory we've used before. It's just-- there's one difference. Namely, we have two optical fields at frequency ω_1 and ω_2 . So for the case of two-photon absorption-- that means both photons are used or stacked up to go up in energy.

We derived this result, and this was the end of lecture last week. And what we obtained in perturbation theory for the excited state, it's exactly the same structure you have seen before. But the only difference is we have now four terms because we have combinations of ω_1 and ω_2 , or we can take two photons out of the same laser beam.

Just sort of a question, just to sort of indicate to you how many terms you would expect when you do the most basic light atom interaction. Just one photon, a plus a dega. How many terms do you get if you write down the Hamiltonian? No approximation without. How many do you get?

AUDIENCE: Four.

PROFESSOR: Four. You have a plus dega for the electric field, σ_+ plus σ_- for

the atom. And then you have four combinations, two are co-rotating, two are counter-rotating. OK. What we are doing here is second order perturbation theory with two optical fields. If you would not do any rotating wave approximation, how many terms would we get?

AUDIENCE: Eight.

PROFESSOR: Eight? I think it's multiplicative because we have four processes involving one-- oops. Now I'm getting confused. I wanted to say 16, 4 times 4. But now I would say in the first step, which is to the intermediate level, we have four at frequency one, four at frequency, which makes eight. But then I think in the second step you get eight more. So if we don't make an approximation, we get 64 terms. But they're just all combinations, all combinations of frequencies.

Anyway, therefore, I hope you appreciate that I did the rotating wave approximation. I said I'm only interested in the near-resonant terms. And then when we say we want to go up in energy in two steps. We absorb two photons. We don't have any emission of photons. These are all the counter-rotating terms. You have only the absorption of photons, and then we have four possibilities. Yes, I mean, you have a term where two photons are emitted from the ground state. This is sort of now doubly counter-rotatable. We're not going there. It's not adding anything new to it. You know what counter-rotating terms mean. In this chapter on two photons, I'm completely focused on the resonant terms.

But since four terms is still too many, I want to just tell you what is special about two photons. I focus now on a situation, and that's the most common situation in the laboratory, where there is a near-resonant intermediate state and that is sort of now filtering out one of the terms. If this intermediate state is resonant with ω_1 , then we only want to consider now the process where the first step to the intermediate step is driven by the field e_1 and the second step to the final state b is driven by the field e_2 . So therefore, we have only one term, which dominates out of those four, or dominates out of those 64, which we would have gotten without any approximation. It's one term-- the near-resonant term-- which dominates. So that's

what we want to discuss now.

So this is the term we want to consider. And it's the same we've always done in lowest order perturbation, second order perturbation theory, in two steps. It's now in two steps. But if you're asking, what is the transition probability? The transition probability, we have to get the probability to be in the excited state. And then we have the usual situation that this term can be written as the sine squared divided by this, and it turns into a delta function times t . And when we divide the probability, the amplitude squared by t , we get a rate. And this is Fermi's Golden Rule. It is exactly the same you have seen probably more than 100 times.

So therefore, we have now a transition rate, which is Fermi's Golden Rule. This is the delta function. I called it the function f because I want to discuss the spectral profile a minute. But then-- and this is the only difference to Fermi's Golden Rule with a signal photon-- the relevant matrix element is, because we have two steps, is the product of two matrix elements squared for step one and for step two. And because we have an intermediate step, we have to divide by the energy mismatch by the detuning in the intermediate state.

But remember, in the one photon picture, Fermi's Golden Rule is the matrix elements squared times the spectral function. So in this matrix element squared in frequency units, neglecting factors of 2, was the Rabi frequency. So therefore, very naturally, we want to define this as a two-photon Rabi frequency whereas each matrix element here divided by \hbar was the single photon Rabi frequency. So therefore, what we obtained for the two photon processes, we have a two-photon Rabi frequency, which is the product of the single photon Rabi frequency for each step, divided by the energy detuning from the immediate state.

So therefore, our result looks almost indistinguishable from the result built on single photons. We've just cleverly defined our quantities. The rate to go from a to b is Rabi frequency squared, but it is the two photon Rabi frequency. And the delta function is the delta function for the resonance-- the energy difference between state a and b -- but now not just minus ω_1 . It is minus the sum of ω_1 plus

ω_2 because we have stacked up the two photons in the two photon process.

So let me just write that down. It looks like the one photon excitation but with suitably defined Rabi frequencies. So in other words, if you were interested just in the physics of two levels-- Rabi oscillation, you name it-- you can just say the same thing happens. The only difference is that instead of having a coupling directly by a matrix element, we are now coupled by this two photon Rabi frequency. And all your equation, you know, everything-- you can consider line shape, spontaneous emission, saturation-- all the phenomena we have discussed for single photon are analogous. You just have to use the density of state calculated for the two photons and you have to use the two photon Rabi frequency.

OK. So I started out by telling you about two photon processes, two photon absorption. But what is maybe even more important in the way how it is used in experiments are Raman processes. So let me just show what I mean. If you have a state a and b, this can be two different vibrational states of a molecule. It can be two hyperfine states of an atom.

Or if you think about a Bragg process, it could be the same internal state of the atom-- the same hyperfine state-- but with two different momenta. Then it is a Raman process only in the external degree of freedom, in the motion away function. The only change is you change the momentum. We need our intermediate state, which is often an electronically excited state. We are detuned. And now we have one photon going up and one photon going down.

Historically, people distinguish between the situation where the final state is lower or higher in energy. One is called the Stokes process. The other one is called anti-Stokes. But as long as you use laser beams to stimulate it, you don't even care which state is higher or lower in energy. But if you work in molecules with a thermal ensemble, then you have certain states thermally populated and others not. And then it makes a difference whether you start from the ground state a or from an excited state b.

OK. So actually, everything for the Raman process is completely analogous-- is

completely covered, actually-- by what I wrote down for you in perturbation theory. It's just if you had kept all the 64 states-- to go up with one photon and down with one photon one was one of them, but we discarded it because we were only interested in going up. So in other words, what was previously one of the counter-rotating terms, where ω_1 had a plus sign and ω_2 had a minus sign, now it becomes a resonant term because we have arranged our two levels a and b in such a way that the near-resonant process is that one.

So in other words, I mention it to you and it's just getting too messy to write it down, when we have $e^{i\omega_1 t}$ and $e^{-i\omega_2 t}$, I mentioned once to you is the sign plus or minus means whether we absorb a photon or whether we emit a photon. If you use a fully quantized picture with a and a^\dagger , the a for the quantized description becomes an $e^{i\omega_1 t}$. In the semi-classical description of the a^\dagger has a minus sign. So therefore, if you look at all the combinations between $e^{i\omega_1 t}$, $e^{-i\omega_2 t}$, for this Raman process we want to select $e^{-i\omega_1 t}$ and $e^{i\omega_2 t}$. And that means we are focusing on this process.

Therefore, everything for the Raman process is analogous to the two-photon absorption process. The only thing we have to do is we have to change the sign on the second frequency because the second photon is emitted in a stimulated way and not absorbed. And therefore, our detuning is the detuning from the Raman resonance. And therefore, OK, back to Fermi's Golden Rule-- the rate in Fermi's Golden Rule is the matrix element squared times the spectral density indicated by the delta function. The delta function is now at the frequency, which is given by the two-photon detuning. And the two-photon Rabi frequency is exactly the same what we had for two-photon absorption, the product of single-photon Rabi frequencies divided by the detuning. Any questions? Yes.

AUDIENCE: [INAUDIBLE]

PROFESSOR: Well, we are usually talking about-- in an atomic system-- about very narrow resonances. And we are working hard on our lasers to be close to one resonance.

And it would be an amazing coincidence if accidentally it would be in resonance with another one. But I would say, if you had a situation-- let's say in a molecule, which is a high density of state, the Raman process would be a rotation or vibration of Raman process involving the ground state. And if you're unlucky and don't choose your lasers wisely, the two laser photons could get you high up into an electronically excited state. And this may have some detrimental effect, depending what you want to do.

But in general, I would say if you have more than one process, there is no interesting interference term. You just get two different rates. One is the two-photon Raman rate and the other one is the two-photon absorption rate. And you just have both simultaneously. They're not leading to the same final state. If something leads to the same final state-- this is more subtle because you kind of cram the interference effects. But we'll discuss some of those things in our next chapter on coherence. Any other questions? OK.

I want to now take it one level higher, where we talk still about two-photon processes but we are allowing one of the photons to be spontaneously emitted. Again, we don't have to learn new things. We just have to map it to knowledge we already have. And let me sort of do it in the following way. I just want to sort of give you a clear understanding what this expression for the two-photon rate is.

If we assume laser one and laser two are near-resonants with a transition a to k and k to b , respectively, we can sort of look at the two-photon process in the following way. We can say the photon ω_2 cannot be absorbed by the initial state. It can only be absorbed by the initial state because ω_1 mixes in with a certain probability, the state k into the ground state. So if you would first forget about it and you just do perturbation theory, then you would say in perturbation theory with the field one, the state a has now a probability, given by this term, that the state a has now an admixture.

And now, if we have sort of-- we have dressed up our state a with admixing for the near-resonant field some probability of state k into it. And this dressed state now

has sort of a stepping stone here. And from this stepping stone, it can now absorb the photon ω_2 . So that's how we should think about it. I'm treating this dressing up of the initial state just a perturbation theory. And that's why everything was in one formula when I applied perturbation theory.

But as we especially cover in the second semester of the course, you can also say if ω_1 is very, very strong, you can exactly diagonalize the Hilbert space of states k and a . And this is called the dressed atom picture. But again, what happens is you mix those two states. And it is the admixture now in a non-perturbative way of state k , which is sort of the stepping stone. And from this stepping stone on, you can absorb a photon ω_2 .

So we could actually-- let me just redraw this-- that we want to go to the final state b . But in this kind of picture I just suggested, I start with a dressed state a . But what is relevant is only kind of this admixture. And from this admixture, we can absorb ω_2 . The real state k is somewhere else. And so it looks like, actually, now a two-level system, where we go from the dashed line-- called the virtual state-- to the final state b .

And let me just point out what this virtual state is. Well, you know already everything about it because everything which can be known about it is what we have derived in our formula. I'm just interpreting the perturbation theory I've written down to you. And if I now call it a virtual state, there is nothing more you can ever know about this state than what was in this formula. But it's maybe helpful to summarize it. Because we have a resonant with frequency ω_2 in this situation, it is clear that the energy of this state is where the dashed line is. It's not the energy of the real state k . The stepping stone is created with the first photon. And the dashed line is the energy level of the virtual state.

What is its character? Spatial wave function and such? Well, it is exactly the intermediate state k . And what is the population? If you had a two-level system, we sort of start with 100% amplitude in state 1. But here, our population is diminished by the probability at which we have it admixed the state. So in other words, if I really

wanted and I want to use this concept, I could make a simplified description of two-photon process. I would just say the two-photon process is just a similar photon process starting from the virtual state, and the virtual state is created by the first photon.

Well, you would say, well, why do you do that? I think the picture I've just drawn for you is sort of helpful when we discuss now two-photon emission-- spontaneous emission. With two lasers, it's sort of simple. But with two-photon emission, we have the situation that we start in an excited state b . We have one laser, ω_1 , and that's it. But we will find out that, eventually, the system populates the ground state.

And one possible process-- and that's the one we are focusing on is-- that it was first emitting a photon in a stimulated way, but the second photon, since we're not offering any extra stimulation, had to be emitted spontaneously. Now, you say, well, how do I calculate it? I can write out long equations. But with the concept I've given you, it should be clear that what we actually have is here, we have nothing-- let's just look at the first part as a dressed atom, an atom in state b with some admixture of state k . And this admixture can now decay by a single-photon process.

So in other words, you don't need to re-derive anything. You can just sort of use analogy to write down what is the spontaneous emission rate out of the state k . So what you would write down now is the rate for this two-photon emission-- one photon stimulated, the second photon spontaneous-- is simply the Einstein A coefficient, or the spontaneous emission rate out of this intermediate state γ_{ka} . But then we have to multiply with the probability that this state is present in the state b because b -- that's what we assumed has no direct matrix element to emit to state a .

And now you should sort of be amazed about the beauty of concepts you have learned. We are talking about something which maybe before this lecture-- wow, one photon stimulated, one photon spontaneous. That must be complicated. But it's just that, except for one thing. And this is the following-- remember, you should always remember how we derived the formula for spontaneous emission. The

physics of spontaneous emission is that you can put one photon in each of the empty modes and you have to sum over all modes. And what was important was the density of modes at the frequency. And now the frequency is ω .

So when we calculated the spontaneous rate-- emission rate-- the decay of the excited state k , we had an ω^3 dependence at the resonance frequency-- at the resonant frequency for the transition k_a . But now we are interested in the density of states at frequency ω . Well, you'll remember, two factors of ω are from the density of states. One came from translating for momentum matrix element to-- no, one was-- yes, it was set. But one was pretty much the single-photon Rabi frequency. And this is also at the frequency of the photon. So you just have to correct for the ω^3 factor. And this is our result now for two-photon emission.

OK. This may be even more relevant at least in the research which is done in my group and in other groups at MIT, are again Raman processes. We often have Raman processes-- you know, you need a reason why you want a two-photon. If you can't reach the upper state with a single photon like people in [INAUDIBLE]-- they may just use two photons. But this is more limit because they don't have the laser, which can bridge the gap. In situations where you work with alkali atoms, we are often very happy with-- we have one resonant line and we can do all the laser cooling, everything we want. But often, we don't like that the line width of the excited state is very large. And therefore, certain precision work, where we want to be very accurate in what we're doing with the atoms, cannot be done on the d_1 or d_2 line. And therefore, we often use a two-level system, which consists of two hyperfine states because then there is no broadening to do spontaneous emission. So one motivation why we again and again and again consider two-photon processes in our laboratory is because it gives us access to very narrow resonances.

But I think you get the gist now, and it will become even clearer later on, that often when you do a transition between two ground state levels, a lot of the physics is the same as of a single photon. You just replace your single photon Rabi frequency by two-photon Rabi frequency, and suddenly you can do everything you ever wanted

to do with a single photon. But now with the benefit of having a very narrow resonance.

So anyway, therefore, I think the most important aspect of two-photon processes 99% of the research people in our field are involved is actually in the form of Raman processes. So therefore, but for pedagogical reasons, I just like to start out with two-photon process-- one photon stimulated, one photon spontaneous. But now let's just fold it over, and we have the lambda type transition. The first photon is absorbed and the second photon is spontaneously emitted. I think as you realize when you go from the two-photon absorption to Raman process, there's nothing you have to re-learn. You just have to be careful with the signs of ω_1 and ω_2 .

So if I would ask you now what is the rate-- what is the rate of the spontaneous Raman process, well, it is the probability to be in the intermediate state. And this probability is just re-writing in a different way what we have used, is the Rabi frequency of the first step squared, divided by the detuning squared. So this is the probability to be in the excited state.

Now, the spontaneous emission occurs with Einstein's a coefficient connecting the intermediate state to the ground state. And then, of course, as we just learned, we have to correct the spectral density and such, or we have to use for the calculation of the density of modes. The correct omega factors-- we have to calculate it at the frequency of the emitted photon. This is actually also-- this kind of spontaneous Raman process-- has been very important historically.

Before the advent of lasers, all you had is light bulbs, strong light bulbs, maybe mercury lamp which put a lot of light into the mercury light, and at least it was somewhat spectrally narrow. But still very, very broad. And you couldn't really resonantly, you know, stack up two light bulbs and have enough spectral power to excite to a certain state. But look here-- you could still, with a strong light bulb, create an admixture of the excited state. This virtual line was terribly broad because of the width of the light bulb-- spectral width-- but then this spontaneous photon was

just compensating for it.

So in other words, it should be obvious-- this process only depends on the power for the first step. And it doesn't really matter if the power is delivered by a laser or a light bulb. The rate for this process is the same. And this actually was the discovery by Raman, which was rewarded with the Nobel Prize, for suddenly observing when you excited molecules with a very strong light bulb, you suddenly saw very different frequencies of photons coming out. And this was a landmark discovery.

OK, what is next? I've already written it out. So that's also important for a lot of research within the CUA. When we simply want to change the momentum state of an atom, we have two lasers. We go up and down. We are not changing the internal state. But it is still a Raman process because state a and b differ by the photon recoil. So we're not going back to the same state. We are going back to the same internal state a, but it may have $2\hbar$ [INAUDIBLE], two-photon recoil different. And therefore, as long as, in quantum physics, one quantum number is different, it is a different state.

And therefore, if you just think about it Rayleigh scattering resonance fluorescence, that you go up and you go down to the next state. You may think you have your favorite atom, and you go up on a cycling transition. Well, when the atom goes up and then emits a photon on a cycling transition, there is recoil involved. So actually what you're doing is on the cycling transition, you cycle it through many, many spontaneous Raman processes. So this is the correct description of resonant fluorescence and Rayleigh scattering. Any questions? Good.

I've just mentioned that when we do Rayleigh scattering, we have to consider that the photons have momentum, and this takes us to another state. Let's now be a little bit more careful and consider what is the role of the momentum in the transition-- in the two-photon process. And in particular, I want to come back to this Fermi's Golden Rule formula and include in this spectral profile, which in the simplest case is always the Lorentzian. But I want to include now Doppler shifts. So in other words, what I want to do now is I want to talk about recoil and Doppler shifts

in a two-photon process and see how it will affect the line shape.

All we have to do is-- or maybe let me back up. You should maybe consider what I've discussed so far, is the situation of an atom which has no motion. We could just fully focus on the internal degree of freedom. And just to remind you, we have discussed two ways how you can eliminate motion out of the picture. One is assume the atom has infinite mass. That's one possibility. The second one is assume the atom is tightly localized in an ion trap, deep in the [INAUDIBLE], that it is localized to less than the wavelength.

And actually, the two ways of how you can eliminate recoil in Doppler shift are actually the same. When I said, give the atom an infinite mass, well, you give the atom an infinite mass by tightly connecting it to the laboratory. And this is what tight confinement in an ion or atom trap does. Because then the recoil is absorbed no longer by the atom, but by your experimental structure, or by your whole laboratory, or even by the building, if you want.

But now, we are kind of going beyond this restriction. We are now saying, OK, now we allow the atom to move. And we can deal with that by simply saying when the atom has a velocity v , we can transform-- we can just use the Galilean transformation and say OK, the physics is the same. However, the atom, due to its velocity, sees a slightly different frequency. So therefore, we have our Lorentzian. But now, we calculate our Lorentzian by using the frequencies perceived by the atom.

The different signs-- plus minus-- are, of course, distinguishing whether we have two-photon absorption or Raman process. And the frequency shift, going into the frame of the atom, gives us Doppler shifts $k_1 v$ and $k_2 v$. And now you'll see that there is something which is potentially interesting, and I want to discuss that. If k_1 and k_2 end up here with a minus sign, it may eliminate Doppler shifts, maybe even completely. So this is something new. If you have a single photon, you always transfer momentum to the atom. But if you have two photons, the two momenta can cancel. And this is actually a powerful method to avoid Doppler broadening in

spectroscopy. So let me elaborate on that.

The message you get from this formula-- it's actually much easier to say it in words than to write it mathematically because it's all hidden in the plus minus sign. But the gist is that two-photon absorption or Raman processes-- so any kind of two-photon process-- they are like single-photon transitions. And we've already discussed it, that we said we have Fermi's Golden Rule like for single photon. It's just we have to use the suitable definition of the two-photon Rabi frequency.

And we had also seen that we can treat, actually, the two photons at one. But the frequencies, sort of, of this photon-- if you want to regard the two-photon process as a super photon, the frequency of the super photon is now either the sum or the difference of the two frequencies, depending whether we look at two-photon absorption or Raman process. But what we see now from the Doppler shift formula, that we can apply the same also to the momentum. It is as if we had a super photon, which drives one transition, but the momentum of this super photon is now the sum or the difference of the two momenta of the photon.

And if you wonder, how do you sum them up? What really matters is what is after the two photons have been exchanged, what is the total momentum transferred to the atom. So what appears here-- k_1 plus minus k_2 -- is the total momentum transfer to the atom. And you see that if you have two-photon absorption, if the two laser beams are counter-propagating, the total momentum transfer is zero. But if you have a Raman process where you absorb one photon and then emit it, the momentum transfer is zero, assuming similar frequencies, when the two Raman beams are parallel.

So in other words, the situation without momentum transfer-- and you will see in a moment, or I've already said this is a situation where you are Doppler free-- for two-photon absorption the geometry is counter-propagating. For Raman processes, it is co-propagating. Then you have no or minimal momentum transfer to the atom. OK. So this total momentum transfer is minimized for k_1 equals minus k_2 for two-photon absorption, or for k_1 equals k_2 co-propagating laser beams in the case of the

Raman process. Oops. Now you have it twice.

Let me just focus, because there is very special interest in that, for precision spectroscopy of hydrogen. Let me assume we have just one laser, which produces - but we want to drive a two-photon process with it. In this situation, ω_1 equals ω_2 . The momentum transfers are the same. And if we arrange for the two photons to be absorbed from opposite directions, we reach the situation where the Doppler shift is really zero. So this is the way where we do Doppler free spectroscopy. And two-photon spectroscopy is one of the handful of methods of practical importance for eliminating the first order Doppler shift.

So if you take an atom-- and let me just quote Dan Kleppner research, where this is the hydrogen atom-- and you have two laser beams from opposite direction. How will the spectrum look like? Well, we have the feature, which I just emphasized, that the two photons-- one is absorbed from the left and from the right, and therefore you get a very, very sharp line. Really sharp. I will talk about it in a second.

But you cannot, of course, suppress the process where you absorb two photons from the left or two photons from the right. And therefore, you have sort of a broad pedestal. So the pedestal is where you take one photon-- both photons from the same side, whereas the Doppler free peak is where you have photons from counter-propagating directions. Since hydrogen is of methodological importance, measurements of-- fundamental measurements of-- the Lamb shift, comparisons with QED calculations, measurement of the Rydberg constant-- these are all done by hydrogen spectroscopy. So therefore, it is very important to have precision method which suppresses the Doppler effect.

However, let me point out that once you have completely eliminated the first order Doppler broadening, you are then limited by the second order Doppler effect. And as I pointed out, there is no geometry-- no arrangements of beams, no tricks you can play-- because one contribution to the second order Doppler effect simply comes from time dilation, that in the frame of the atom, relativistically speaking, the clock ticks differently. And therefore, the spectral line is different.

So this is what we had discussed already for the second order Doppler effect, and this is not suppressed by two-photon. If you now estimate what is the relative line width-- so what is the delta, the line broadening you to the second order Doppler effect, in relation to the transition frequency? So just give me a second. Yeah. Then this omega cancels and we have an expression which is sort of interesting. It is mv^2 . It's the energy-- the thermal energy-- but since we have normalized it to the transition frequency, it becomes now the thermal energy relative to the rest mass of the atom. You would think, well, this must be really tiny. Well, it is tiny.

If you take room temperature, it is 2×10^{-11} . But people are now looking for precision in optical clocks, which is in the 10^{-15} range. So you really have to be able to know the temperature, know the kinetic energy distribution of the atom, and be able to correct for it. Or ultimately, if you can't correct for it, you would not be able to correct for it at room temperature because you can't know for certain that the velocity distribution of the atoms in your laser beam is exactly at room temperature. You really have to go to low temperature, go to cryogenic temperatures.

And in the famous experiments in Munich and [INAUDIBLE] group, the typical situation is for the 1s to 2s transition, they observed residual line broadenings on the order of a few hundred Hertz. This is not limited at all by the natural lifetime of the 2s transition. As you calculate in your last homework assignment, the lifetime of the 2s state is actually due to two-photon emission. Amazing. Two photons in series. This is the way how the 2s state decays.

And you will, actually, with this rather simple description, get a fairly accurate estimate for the lifetime, which is a fraction of a second. And [INAUDIBLE] group uses a cryogenic experiment. They cool the hydrogen by collisions with liquid helium, cooled vaults, to maybe a Kelvin or so, three hundred times below room temperature. And this has been important to reach this precision. And still, I think, even at Kelvin temperature, the second order Doppler shift is one of the important-- is one of the important systematics.

OK. This is what I wanted to discuss with you about two photons. Any questions?
Yes.

AUDIENCE: Just-- very lightly-- I would have thought [INAUDIBLE].

PROFESSOR: That's correct. It really depends what you want. If you simply want an atomic clock, all you want is a very, very, very stable reference point. And people use for that [INAUDIBLE] and strontium, or the people who do ion traps use the aluminum ion. All they want is a spectral line, which is sufficiently sharp, sufficiently narrow, and also insensitive to magnetic fields and electric fields. And so they select all over from the whole periodic table what they want.

But people who want to measure fundamental constants-- the Rydberg constant-- want to compare lame shift with first principle QED calculations, sort of test the precision of quantum electrodynamics, verify that quantum electrodynamics is a complete description of atom photon interaction, they can only deal with simple systems where all the calculations are possible. This is the case for hydrogen.

And, actually, with some advance in the numeric calculation of wave functions and all that, it may also be possible to do it with helium. I know in the literature people have often suggested helium have pushed the precision of two-electron calculations further and further. I think so far it hasn't kind of-- helium has not replaced hydrogen. It's still hydrogen. But this sort of tells you what the choices if you want to test fundamental physics or determine fundamental constants. In all atoms other than helium and hydrogen, you would be limited by the infeasibility of many electron calculations. Yes?

AUDIENCE: Going back to the two-photon Raman process where you had the second spontaneous emission, I just want to clarify. So in the past, all this time when you were talking about off-resonant single photon scattering, was this actually, really, the more descriptive picture or was there actually a different physical--

PROFESSOR: No, this is off resonance scattering. And if you ask me, when do you have a situation where you first absorb the photon and then emit it, I would say, I would like

to know that. I don't think there is a situation where this is possible. You always-- you should always use a two-photon picture. Or the only situation, if you press a little bit harder, where I think you can think in first absorption and then emission, is if you have a gas with lots and lots of collisions, the first photon may not be fully resonant. But then you have a collision and the collision stabilised, provides the missing energy or takes away the extra energy.

And then with something which is much more complicated than you ever wanted, you have now truly an atom in an excited state, which has completely lost its memory from how it was excited. And yes, for this atom, it will now simply spontaneously emit a photon. But in all other situations where you don't have any loss of coherence or something in between, you're never really going to the real state. You're always going to virtual state. It's all two-photon. And, I mean, we discussed that.

Remember the clicker question where most of the class was confused when I said we go up-- if we're exciting the atom? But now we are asking, what is the spectrum of the emitted photon? It is a delta function at the drive field, and we discussed it at length. If you allow in your head any picture of first absorption then emission, that's where all the wrong answers come from. This is really the picture behind it. This is what happens. Photon in, photon out-- should be described together, unless you have-- and I'm just repeating myself-- something in between, which is sort of some form of de-coherence which decouples the two processes.

OK. But that's a wonderful opening to our next chapter, namely coherence. To what extent-- we just discussed one aspect of it and we come back to this in this chapter-- to what extent is the photon which is scattered coherent with the incoming photon? So I want to feature, in this last big chapter of this course, coherence in all its different manifestations. I think this is rather unusual. I don't know of any textbook or any other course where this is done. But this is similar in spirit to what we did on line broadening. I felt I could create spatial connections by discussing all possibly line shifts and line broadenings together. And now I hope you will also see certain common traits if I discuss together all the different manifestations of coherence.

So what we want to discuss is-- we start out by talking about coherence in a single atom. We can have coherence between two levels. Usually, I try to stop at the simplest possible manifestation. But when we talk about coherence, I cannot stop at two levels because there are many new qualitative features which come into play when we have three levels-- like lasing without inversion, like electromagnetically-induced transparencies, for those of you who have heard about it. On the other hand, I can reassure you, I don't think there's anything fundamental to be learned by going to four, five, and six levels, so we will stop at three levels.

We can have-- so this is a single atom. But we can also have coherence between different atoms. And phenomena we want to discuss is superradiance, which is very, very much related to the phenomenon of phase matching. Everybody who frequency doubles a laser knows about phase matching and phase matching conditions. You have to rotate the crystal or heat the crystal to the temperature where the whole crystal-- all the atoms in the crystal act coherently. But it's very related to superradiance.

There is a third aspect of coherence between atoms which I will not discuss this semester, and this is the situation of Bose-Einstein condensates and macroscopic wave functions. This is covered together with quantum gases in-- this is discussed in the context of quantum gases in the second part of the course.

So having these very different manifestations of coherence, I want to try now to give you a definition of coherence. But it's a bit difficult because I want to cover with my definition all the cases I know. But with those examples in mind, for me coherence-- we have the phenomenon of coherence. Coherence exists if there is a well-defined phase. Well, if we have a phase-- a well-defined phase-- it's always a phase between quantum mechanical amplitudes. So we need two or more amplitudes.

So coherence exists if there is a well-defined phase between two or more amplitudes, but we can only observe it if those amplitudes interfere. And it can be two amplitudes describing two different atoms, or it can be two amplitudes of two states within the same atom. But I will point out to you-- what is really relevant is an

indistinguishable ability that those two amplitudes are involved in two branches of a process which has the same final state. And like in Feynman's double-slit experiment, you don't know which intermediate state was taken. And that's where coherence manifests itself.

So that means when we observe an interference, that means we observe-- and this is how we read out of a coherence-- one observes a physical quantity, the population in a certain quantum state, total electric field emitted, but this quantity is usually proportional to the square of the total amplitude. And that means we get an interference term. So coherence is important.

Let me provide one additional motivation that coherence is an important technique and important tool for measurements. In a way, it's subtle but trivial at the same time. Whenever we do spectroscopy, we are actually interested in doing a measurement of energy. We want to measure energy levels. And those energy levels can tell us something about magnetic fields through Zeeman shifts. If you're addressing energy levels in the gravitational field for atom interferometry, the energy levels reflect gravitational fields. Or if we are not interested in-- if we try to eliminate or shield the atoms from magnetic fields and we just want to get the most precision in a reproducible energy level, this is the situation of atomic clocks.

So pretty much when we use atomic spectroscopy for any application, we are interested in the energy levels. But this is very deeply connected to coherence and the phase because the relative phase between two states is nothing else than the time integral over the energy difference between two levels. So the phase evolved between the two levels is the difference frequency times time or time integrated dt .

So therefore, when we are talking about coherence, how can we maintain longer coherence between energy levels or how can we create coherence in three level systems? This is actually intricately related to the fact that we can obtain more precise information about the energy levels. Anyway, this is a very general introduction to coherence. Before I talk about manifestations of coherence, I have two clicker questions because the first form of coherence I want to discuss is the

coherence of-- the coherence involved in exciting atoms and the atom emitting light. It's related to the spontaneous emission and scattering problem.

So just to sort of figure out what you know already, let me ask you something about the nature of spontaneous emission. The first choice is spontaneous emission, is nothing else than a unitary transformation-- unitary transformation or unitary time evolution-- of the wave function of the total system. And option B is spontaneous emission introduces-- well, maybe through a master equations or optical Bloch equation-- introduces a random phase into the time evolution of the quantum mechanical system.

So what is the picture you have on spontaneous emission? OK. Stop. OK. So at least half of you better pay attention now. So the answer is whenever you have a system and it couples to the electromagnetic field-- you just put a system in an excited state and you wait. The 100% unique and correct answer is the system involves with the following operator, and this is the operator we have discussed many times.

This is the operator which completely describes the interaction of an atom with the electromagnetic field. And since the whole system is completely described by this Hamiltonian, the whole system undergoes a unitary time evolution. So if you talk about the total system consisting of the photons in all electromagnetic-- in all relevant modes-- and the atomic system, this total quantomechanical system has a unitary time evolution with this operator. And to the best of our knowledge, this is a complete description which covers all aspects of the system. OK.

But, and this is now the next question, there is a certain randomness in spontaneous emission when we go to the laboratory and look at the spontaneously emitted photons. And this is actually what I want to work out with you in-- maybe even today, I think ten minutes may be enough-- what is really the information-- the phase information-- which we have in a photon, which has been spontaneously emitted. I know how to phrase-- the randomness of spontaneous emission. Well, let me write it down.

First a very big disclaimer. This question does not contradict the first one. The fact that we have a unitary evolution with this operator is 100% or 110% true. But this operator will actually lead to final states of the photon field, which may not have a specific phase. So if you say there is some intuition that there is something going on with the phase, you may be correct. But everything which is going on with the phase is the result of a unitary time evolution. The system itself is described by an operator, by a Schrodinger equation for the total system.

But the question I have for you now is if you detect, let's say, the photon emitted in spontaneous emission, the randomness of spontaneous emission, the-- let me call it loss of phase, or at least the diminishment of the read out. There may be situations where we have a laser beam which has a well-defined phase, photons are scattered, and we just cannot retrieve the phase of the laser beam by looking at the photons. So this is what I mean here. Also, the photons can't come out of a unitary time evolution.

My question now is, is the randomness or this loss of phase of spontaneously emitted photons-- and now we should, I want to know your best guess-- what is it due to? Is it only due to the-- does it only occur, is it only due to the measurement process of the photon? Or is it due to performing sort of a partial trace ever-reaching over certain states? So if you're interested in the photon, maybe tracing out the states of the atom. Or ever-reaching over modes of the electromagnetic field. And question C is both is actually possible.

So if you look at spontaneously emitted photons and they're not perfectly phased coherent-- they're not reproducing the phase of the laser who has created them-- what is the reason for that? Is it always the fundamental reason or is there no fundamental reason? It's only a kind of reason of ignoring information, taking a partial trace? OK. That's pretty good. Yes.

What I want to emphasize is that A is very, very important, and I want to discuss it now. But B is always the case. If you ignore the position where the atom has scattered the light-- if atoms scatter light and they are wavelengths apart, then you

have maybe optical path length differences. The photon from the laser hits an atom here. It goes to your detector. But from another atom, the photon has accumulated a different spatial phase into the IKR. And, of course, what you get here is the random phase.

This is why quite often when we scatter light for many, many atoms, we're not even asking for the phase. We say, one atom scatters light-- really scatters light at a certain intensity, I_1 . And n atoms-- well, we get n times the light. You immediately perform an incoherence sum because you sort of know deep in your heart that there won't be any interference. So it's always possible, of course, to lose the phase by not controlling every aspect of your experiment. So B is always possible. But for so many reasons that I don't want to discuss it, but the measurement process is very relevant. And so, therefore, I would have answered C here.

But let's now discuss the most fundamental situation. And the most fundamental situation is we take our favorite atom with infinite mass-- no Doppler shift, we just put it. And we put it in a cavity that it can only interact with a single mode of the electromagnetic field. So this is a fundamental situation. And a lot of the general situations, you get by just summing up over many modes, summing up over many positions of the atom, introducing Doppler effects and all that. It just messes things up. And this is more in the spirit of answer B, that you perform partial trace and average over many states. But let's now pinpoint what I think is intellectually the most important one, the pure situation where an atom is just talking to a single mode.

So let us assume that we have an atom which starts in the ground state, but now we excite the atom. And we excite it by a short pulse. And this can be a pulse which has a pulse aimed between zero and π . And depending what the pulse angle is, it will admix-- we have the ground state and it will admix something of the excited state. And in case of a π pulse, we have 100% in the excited state. So our atomic wave function is this.

The excited state-- let's just assume the photons are in resonance. We know how to

deal with off resonant lasers. So therefore, the phase evolution is $e^{-i\omega t}$. But now, and this is what coherence is about, there is a very specific phase. And this phase ϕ comes from the laser. If you excite the atom with a laser beam but it has a phase shift, then the atomic wave function is phase shifted because every amplitude you admixed into the ground state in form of an excited state was driven by the operator-- the dipole operator, e^{+} and e^{-} , the electric field, has the phase of the laser beam. Sure, there may be-- and kind of all other trivial factors I've set to one here. But there is the phase of the laser, which directly is imprinted into the phase of the wave function. OK.

So this is what the atom does-- what the laser beam does to the atom. So we have now an atom which is partially excited. And it carries is an imprint of the phase of the laser. And now after the laser pulse is over, the photonic part of the wave function is a vacuum. We have no photon in our cavity. And now we wait and we allow spontaneous emission. And spontaneous emission is nothing else than the time evolution with the operator I just discussed with you.

So after spontaneous emission, well, one is we know for sure the atom is in the ground state. Let me write down the result. If we apply the operator which couples to the electromagnetic field, and we assume only co-rotating terms here. Let's just neglect counter-rotating terms, which can be-- which in near-resonants are irrelevant. What happens is we are now propagating this wave function.

And the ground state-- with the ground state of the photon, does nothing. However, the excited state with zero photon, we discussed that excited state with zero photon will actually do Rabi oscillation. Ground state with one photon, excited state with zero photons. And so we have now our knowledge from the vacuum Rabi oscillation that this part of the wave function does nothing. This part of the wave function undergoes single photon vacuum Rabi oscillations.

And if we start out with the superposition of ground zero and excited zero, well, this is a superposition principle of quantum mechanics. We can just propagate this part, and we can propagate that part. So what I suggest is when we, really at the

fundamental level now, discuss spontaneous emission, we allow this part-- excited atom, empty cavity-- to undergo half a vacuum Rabi oscillation. And then the excited state is in the ground state and the photon state has one photon.

It's a completely coherent Rabi oscillation. I just allow half a cycle to evolve. And the result of that is, well, nothing happened to this part. And the Rabi oscillations have now taken us to the one photon state. And it has just swapped excited zero to ground state one photon.

So let me write down something which is really remarkable, and then we discuss on Monday-- next Monday-- we discuss how we would really measure the phase. But just look at the two expressions I've underlined. And this tells us that the quantum state of the atom as a two-level system-- ground excited state with all the phase factors-- has been now exactly matched on the quantum state of the cavity. So if you regard ground and excited state as a two-level system, every quantum mechanical subtlety of the atomic system has now disappeared. It's in the ground state. But everything which was coherent, which was a phase which was interesting about the atomic system, has been transferred to the photon field. Yes?

AUDIENCE: Do you want to have an αe there also?

PROFESSOR: Oh yes, please. Thank you. When I said everything, I meant everything. Yes. Yes. OK. So let me write that down and then we have-- so this is what I meant. This is the most fundamental aspect of spontaneous emission, that the quantum state of the atom has been perfectly matched. Perfectly mapped onto the photon field.

And the one thing we have to discuss on Wednesday is the whole of the phase ϕ . ϕ -- we started out by ϕ being the phase of the laser. And if the laser is in a coherent state, I will talk about on Wednesday, in a homodyne measurement we can measure ϕ to any accuracy you want. We can determine the phase of the laser in a homodyne or heterodyne experiment. This phase ϕ has been now perfectly imprinted into a two-level system for the atom. And now it appears mapped into a two-level system for the photons-- the two-level system between zero photons and one photons.

But if we are now doing a measurement either on the atomic system or on the photonic system, we are limited in the accuracy at which we can retrieve ϕ . And this is what we want to discuss on Wednesday. And this is what I referred to as the fundamental limit of spontaneous emission because we have not lost any coherence here. It's just if the phase is only imprinted in one particle-- one particle quantum physics sets us a limitation. Oh well, we can read out the phase ϕ . OK. Any question? To be continued on Monday.