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I was invited to talk here about entanglement optimization in plasmonically coupled quantum dots. So that is the title of my talk. As a advanced warning, I'm going to talk about a bunch of stuff related to this. But I promise to have at least one slide on entanglement optimization in plasmonically coupled quantum dots.

As another advanced warning--

this is almost exclusively theoretical work. So the experiment is a work in progress at this point. And the theory has been in collaboration with Stephen Gray and our students and post-docs at Argonne National Laboratories. So he deserves at least as much credit for this as I do, but I'm here, so [INAUDIBLE].

So to go back to how we got into this, our motivation for starting to look at quantum dots coupled to the plasmonic systems. So plasmons, plasmonic metal nanoparticles have been looked at for about 40 years. And most of that time the motivation has been for enhancing optical processes. So starting with enhanced Raman scattering. And you can enhance almost any optical process because of the various strong localized fields that you get around metal nanoparticles when you excite plasmons collective electron oscillations inside those particles. And so optical emission light emission from a luminescent object is another process that can be enhanced if you put the object next to a plasmonic metal nanoparticle.

And since this has been studied by a large, diverse group of scientists over a long time, there have been a lot of sort of descriptions of this process that have come in. So if you're a chemist, you probably like to describe it through an energy transfer process, where the molecule or atom or whatever is emitting light transfers energy to the plasmon resonance in the nanoparticle. And then that nanoparticle either absorbs light, or radiates it to the far field.

If you're an electrical engineer, and you like to describe it in terms of an optical antenna, the metal nanoparticles, the optical frequency analog of the antennas in your cell phone which take the radiation from this small dipole, and transmit it to the far field in the same way that the antenna in your cell phone radiates the signal from the oscillating current inside the [INAUDIBLE] circuit to the far field, but at much higher frequencies.

And then if you're a physicist like me, you like to think of this in the cavity QED pictures. So the thinking as the metal nanoparticle as the antenna, you think of it as a resonant, as the equivalent of the optical cavities that you heard about in the last two talks, by taking an emitter, like a quantum dot or a molecule, and putting it in the near-field of the plasmonic cavity, you can enhance the spontaneous emission rate by enhancing the available density of electromagnetic bonds so that we can emit more quickly into the plasma and resonance frequency the same way as a [INAUDIBLE] inside a photonic crystal cavity or a micropillar cavity can emit more quickly into that cavity mode, and then that cavity can transmit that to the far field or not depending on its properties.

Of course, all of these are equivalent descriptions of the same underlying physical processes, but they give you a slightly different perspective on what's going on. In the cavity QED perspective in particular, we [?] may?] start to think that maybe we could do things by calculating quantum dots or other emitters to plasmonic particles that are analogous to what you do in cavity QED when you increase the coupling strength between the emitter and the cavity. This enhanced emission is what we would call weak coupling cavity QED, where you're taking the spontaneous emission process that have already happened, and just increasing the rate at which it happens.

But over a lot of time, the sort of systems that we heard about in the last couple of talks, people have gotten into

the strong coupling regime, where the emitter and the cavity mode are no longer separable. They form new coupled states. And you get strong modification of the spectral properties of these things, and the potential for a quantum state manipulation.

So since the same physics is there in the plasmonic systems, of course the parameters regime is quite different. Plasmons in particular have very short lifetimes. They decay and dephase. They tend to femtosecond time scales. So the question of whether we could achieve these same sort of strong coupling effects or analogous effects in a plasmonic system is at least not initially completely obvious.

And so because of those sort of the decoherence times, we're probably not going to use these for the quantum information processing applications that are used in photonic cavities. But we can, in principle, still get into regimes where there are strong interactions between a single quantum dot, and a plasmonic system that can lead to strong modifications of the optical properties that go beyond sort of a simple combination of the properties of the quantum dot and the plasmonic particles themselves.

So going back several years ago, we decided to try to make a realistic calculation--

"realistic" meaning realistic parameters for the plasmonic metal nanostructure for the quantum dot that was coupled to it. And so this is the system of a pair of silver nanorods with a single quantum dot in between it. And the quantum dot we considered was not the indium arsenide [? gallium ?] arsenide would be surface grown [INAUDIBLE] quantum dots you've heard about in the last couple of talks, but the colloidal quantum dots, are a little spherical particle of cadmium selenide of the kind that you can make using [INAUDIBLE] chemical synthesis methods.

We saw that if you look at the scattering spectrum of the plasmonic nanostructures themselves, the green dots, and you put a single quantum dot right where this field is the strongest between the two of them, you can get nearl

y complete cancellation of the scattering, and the absorption of the much larger plasmonic metal nanostructure. And this happens partly because of strong coupling, with the normal mode splitting between the coupled [INAUDIBLE] quantum dot dipole and plasmonic dipole.

But also it's dominantly an interference effect. Because the quantum dot is so strongly coupled to the plasmonic system, it can nearly cancel out the absorption of that system through this coupling, even though its own dipole excited by the far field is so much stronger-- so much weaker.

So this is analogous to the similar effect that was actually [?] connected [?] by [INAUDIBLE] several years ago for quantum dots inside photonic cavities. So what he called dipole-induced transparency, we call this quantum dot induced transparency. In the plasmonic field, we probably should have called it a Fano interference effect, because this became the preferred language for these sorts of interference-induced spectral modifications.

So people have for several years seen these in plasmonic systems by coupling different modes of plasmonic nanoparticles together, coupling a bright mode that radiates to the far field, and a dark mode that doesn't couple, or couples weakly to the far field, you can get the similar sort of interference effects going on.

But those spectral responses are fixed. That's just set by the geometry of the nanoparticles. Our interest here was that this is something that in principle you could turn on and off optically.

A single quantum dot has two excitons in the ground state, and one's spin-up and one's spin-down. So if you excite a single photon inside the quantum dot, the quantum dot should become transparent. And this induced transparency dip should disappear. So just absorbing one photon inside the quantum dots should create the very large modulation of the cross-section of this coupled system.

So the model of this, we went all out and did a full quantum mechanical model. We quantized the plasmon fields and the state inside the quantum dot. We coupled the two of them together. Looked at that, coupling damping of the two of them and solved the master equation, and through all of that work showed that as we increased the intensity of the light that's incident under the system, this induced transparency dip indeed does get smaller and smaller, and eventually disappear.

Because we had all these equations that we were solving in the time delay, we could also look at what happened if instead we should send in short laser pulses, instead of just a continually shining light on it. And here we could see that not only does the dip disappear, but at particular intensities of this [INAUDIBLE] energies in these incident pulses, the sign of the interference actually reverses, and you go from having induced transparency into this super scattering spike to induced additional absorption.

So you're changing the interference from destructive to constructive. This happens because the instant pulse is driving the quantum dot through a full Rabi oscillation. So it's going all the way into an excited state [INAUDIBLE] back down to the ground state, accumulating a [ $\pi$ ] phase in the process, and thereby changing the interference between it and the plasmonic dipole.

So all of this, well, it was interesting to see it show at least in principle that we could build ultrafast switches out of these plasmonic switches, or moderators, shall we say, something where you can use an optical signal to control another optical signal. It should happen on very short timescales, on the femtosecond time scales, because these are the coherence times [INAUDIBLE] systems. In principle, it should be a very low power modulator, because it requires only a single photon to be absorbed by the quantum dot in the system, and this on the nanometer scale. So those are really the ingredients that are needed for the optical information processing systems that

that will ultimately be needed to overcome the sort of power energy constraints that are limiting on-chip or chip-to-chip data transmission in microelectronic systems right now.

But this is all really classical, or semi-classical. even. Though we went through the trouble of modeling everything quantum mechanically, the interference itself, you can understand that purely classically. It's an interference between two classical dipoles.

And even this disappearance or reversal of the peak, we could model using a pretty simple semi-classical model. So this is its own line is the semi-classical model as compared to the full-quantum model. Apart from some small numerical differences, they really are equivalent to one another.

So we wanted to see is there anything that can happen in the system that is really quantum mechanical in nature? And the one thing is indisputably quantum mechanical is entanglement. There is no classical analog of quantum entanglement.

It seems that entanglement should be very hard to create in the system because of the very strong dissipation that goes on. Well, it turns out that under the right circumstances the dissipation itself can serve as a tool to create entanglement in the system. So if you imagine we go up just a little bit in scale, now we have two quantum dots coupled to a plasmonic system-- so a system of coupled metal nanoparticles. And we start with one of those quantum dots in its excited state, and the other one in the ground state.

Then what happens is they, on a very short time scale, that initial state evolves into an entangled state of the populations of the two quantum dots. And if we look at the concurrence which is a measure of the degree of entanglement between the states of the two quantum dots, it rapidly reaches a value that indicates some entanglement, then decays away as the population of quantum dots decay away.

So what's going on here is the initial state, the state of one excited and one in the ground state. You can see as a superposition of symmetric and antisymmetric states of  $1 + 1$ , and  $1 - 1$  as the quantum dots.

And the symmetric state couples strongly to the plasmonic system. It's a superradiant state, whereas the antisymmetric state is these are the analog of a triplet state that's a sub-radiant or a dark state. And it couples weakly to the plasmonic system. So it's the [? plasmonic ?] damping by the plasmonic system of the quantum dots that removes the symmetric state, leaving behind only the antisymmetric state, which is an entangled state of the two quantum dots.

So this is something that around the same time was predicted by the group of Garcia Vidal for quantum dots coupled to plasmonic waveguides. We decided to look at whether we could scale this up to multiple quantum dots. So if you have, again, you take several quantum dots-- three of them, four of them, or more-- couple them to a single plasmonic system. And the idea here is this is all something that can be done on the sub-diffraction limited scale.

So in the near field, now we're not talking about sending photons back and forth along the wave guide between these things, but just having a system where you start with a population of one quantum dot in its excited state, and all the other ones in the ground state. We would again get pairwise entanglement between that initially excited quantum dot, and each of the other quantum dots inside the system. And that entanglement induces entanglement between the quantum dots that weren't initially excited as well, although to a lesser degree.

What we've taken here is the difficulty of creating an entangled state, and translating to the difficulty of somehow exciting just one of these quantum dots that are all right next to each other into its excited state initially. So I wouldn't say we're trying to get experimentally realistic, but slightly more realistic.



c. We want to see if we could build a system where the ability to create entanglement just came into the design of the system, of these metal nanoparticles, of the quantum dots that are coupled to them. And we could just shine the laser pulse on it, and create an entangled state through the natural evolution of the system. So sort of spontaneously through the coupling to the plasmonic system.

So the idea that we had was we could vary the coupling strengths between each quantum dot and the plasmonic system. So vary the degree to which a quantum dot dipole is coupled to the plasmonic dipole, which in principle, you could do just by changing the separation between the quantum dot and the plasmonic particles.

So if you have a quantum dot between two metal nanoparticles, you move the metal particles closer together, the coupling will be stronger. You move them further apart, the coupling will be weaker. So by having multiple quantum dots in the gaps of different widths, we could tune the couple strengths for each of the quantum dots individually.

And so our initial thought was we excite the whole thing with a laser pulse. The ones that are more strongly coupled will get excited more, and the ones that are more weakly coupled will get excited less. It actually turned out to be kind of the other way around. And it worked best, if we excite the system very strongly, we can make one of the quantum dots undergo a full Rabi oscillation. And so it goes from the ground state all the way back down to the ground state.

And say another quantum dot go through half a Rabi oscillation from the ground state to the excited state. So you have one that go through a power of  $\pi$  then go through a  $\pi/2$  pulse. So you create the initial state that you want by, in this case, cha

changing the structure, rather than changing the input fields to the system. So if addressing you to the quantum dot separately, you address them all simultaneously, but you build your structure so that they'll end up in the state that you'll want.

And then that initial state evolves and produces the entanglement that you want. Entanglement is a little bit lower, because we don't have exactly the same decay rates. We don't have a pure symmetric and antisymmetric state right now, because the two quantum dots are no longer equivalent to one another.

Well, actually turns out that having completely equivalent quantum dots is not the optimum in terms of getting the highest degree of entanglement. So the highest concurrence numbers happen when the ratio of the coupling strengths of the quantum dots,  $g_2$  is the strength with which one quantum dot is coupled to the system, and  $g_1$  is the coupling strength for the other one. You can show analytically that they have a coupling strength ratio of [INAUDIBLE] 3. Then, an initial state with one quantum dot excited, and the other one on the ground state, will evolve to a state with the greatest degree of entanglement. This is the optimization side [INAUDIBLE].

So that's one optimization condition. The other optimization condition is that you want to create, as close as possible, the state in which only one quantum dot is initially excited. And so there you want one of the quantum dots to go through integer number of Rabi oscillations, and the other one to go through a half-integer number of Rabi oscillations.

So if one goes through a half, the other goes through one, or one goes through three halves, the other one goes through two. Either way, the ratio of the two of them should be given by the ratio of an integer divided by a half integer.

Of course, you can't reach both of those at the same time. So the rate of entanglement you can get is given by how close you can get to satisfying

those two conditions simultaneously. The 400 doesn't seem to work anymore.

So the graph shows the brute force calculation of all of the different coupling strengths, and all the different pulse energies that you might want to put into the system. And you can see there's a bright spot right in the middle there. The dotted line is this condition. And so just a little bit below it, we have this condition. This is the best. But we also got maxima close to other places where this condition is fulfilled.

So we have a procedure that we can use to optimize the design of our system to get the most amount of this transient entanglement among the quantum dots that are coupled to it. And these are analytical formulas. And we can easily scale them up. The equations get a little messy. But the idea is very much the same. We can entangle three or four or more quantum dots simultaneously by tuning their individual coupling strengths to this plasmonic system.

So, four minutes. To say there are two things that you might ask at this point. The first one is, can we actually accomplish this experimentally? The other one is if we can accomplish it experimentally, is it good for anything?

The first question is a work in progress. But we are working at least towards the first stages of this, of taking individual plasmonic metal nanoparticles that are synthesized kind of within solution, and binding individual quantum dots onto their ends by using selective chemical methods. With this chemical synthesis, we can control the separation between the dots and the plasmonic metal nanoparticles by the lengths of the molecules that we use to bind them to one another. So we can have very precise control over this coupling strength.

Between them, this is, in principle, also something that's scalable. We can put one dot

on. Then we can put another metal nanoparticle, and another quantum dot, and build this up. We're working on the first stage of this right now.

To answer the second question, I'm not sure whether we could do anything useful with this in the quantum information sense. From our point of view, or from my point of view, I should say, this sort of satisfied me that quantum mechanics --

-- real quantum mechanical effects are at least not incompatible with the ultra- fast dissipation that you have in the plasmonic systems. But it also shows that we can engineer the systems and their optical excitation to coherently control the optical energy and the states that are inside the system.

Through this reversal of the transparency peak in the coupled quantum dot system, we showed that we can use external fields to coherently control the collective response of the system. And then this entanglement work, we showed that we can engineer the structure of the system to drive this system to evolve into a desired final state just by changing its structure, not by externally controlling it.

So if we generalize this, we might be able to find principles for designing these coupled plasmonic quantum dot systems to get full spatial temporal control over energy and excitations on the nanometer-length scale and on the femtosecond time scales within these systems. So for example, you can imagine putting together a system of metal nanoparticles with different dimensions to coherently absorb up the solar radiation over a broad bandwidth, and over a large absorption cross-section, and then concurrently transfer that energy to a single quantum dot that you could use for solar energy harvesting in an efficient [INAUDIBLE] system.

I should be done now, right? Two more minutes? All right.

So to go from that, since I mentioned solar energy, I will show a slightly related idea that comes from the same calculations. When I mentioned this idea of coherent solar energy harvesting, people say, of course, there are other ideas out there for using plasmonic systems to enhance the efficiency of solar cells. So this, from a few years ago now, or so was summarized in a review paper from Harry Atwater and Albert Polman.

And to make a long story short, two out three of these methods are really far field methods. Both of them are actually based on increasing optical path-lengths inside the photovoltaic or solar energy conversion device through multiple scattering or through coupling to waveguides. In those cases, you probably would be better off using high dielectric constant-- dielectric particles that don't absorb a lot of light, rather than introducing metal that absorbs a lot of light into the system.

The only way you can really take advantage of the plasmonic resonances per se is through the third scheme, which uses the near fields to locally enhance absorption in the nanometer-scale volumes next to the [INAUDIBLE] particles.

Of course, absorption is a linear process. If you have a higher field somewhere, you have lower field somewhere else. Energy is conserved. So averaging over the volume of the solar cell, the total amount of absorption inside the active material can't be increased by shifting fields from one location to another, unless the solar cell itself is very thin compared to the volume of those metal nanoparticles. So the plasmonic local field enhancement is only going to enhance overall absorption in your solar cell if you're talking about solar cells with active materials thicknesses on the single nanometer scale.

There are a couple of systems where you might want to do that. So for organic photovoltaic dye-sensitized solar cells, transport through those materials, through the light-absorbing active layers, is so poor that you might want to make those active layers as small as possible.

So we use this quantum model that we had to model a dye-sensitized solar cell to make a very simple toy model of a dye-sensitized solar cell, which is a single dye molecule coupled to a semiconductor like titanium dioxide, and plasmonically enhanced by coupling that dye molecule to a nearby plasmonic metal nanoparticle. So the [? nano ?] nanoparticle can enhance absorption by the dye molecule, and that dye molecule transfers an electron to the semiconductor. And the rate at which it transfers electrons to the semiconductor is a proxy for the photocurrent that would be produced in the solar cell.

And we saw the black dots correspond to the more realistic system parameters that we simulated. As you increase the coupling strength between the dye molecule and the plasmonic metal nanoparticle, you can enhance this photocurrent up to a certain point. And then you actually start to decrease the photocurrent that you produce again, looking at both for different plasmonic relaxation times for different electron transfer times to the semiconductor.

And so the reason for this is that the Purcell effect that I mentioned at the very beginning of the talk, this enhanced emission, if you had enhanced absorption, you necessarily enhanced emission or relaxation by the dye molecule as well. And eventually, that relaxation becomes faster than the electron transferring to the semiconductor. You might absorb more light in the dye molecule, but you don't get that out as a current in your photovoltaic in the end anyways. So the somewhat pessimistic message of this, is you can do well, but you can't just keep increasing your coupling strength, and hope to keep doing better and better. There really is a limit on how much more you can get out using plasmons.

So with that, since you've been standing there for a while, I'll finish. Happy to take questions.

I don't think we have time for--

[INTERPOSING VOICES]

We need to pass directly to the next session. We announce the best paper award [INAUDIBLE].

[INAUDIBLE] conference. [INAUDIBLE] competition. The competition had two phases. The first phase is [INAUDIBLE] by our program committee. And then the strongest talks were chosen to be evaluated here by the viewers on site for the best [INAUDIBLE] presentation.

We had so many [INAUDIBLE] fantastic talks from students. And it was amazing that [INAUDIBLE] contributed in the conference. And it was really very, very tough choice to select. Given that there's such amazing strong contributions, we will give the awards this year for third place, second place, and first place. And there is also based on the ratings from the jury.

So I would like to call first the third place talk for the best student paper award. And this is by Sophie [INAUDIBLE]. And the title of the talk was [INAUDIBLE] Based on the Transfer Electromagnetic Angular Momentum." Congratulations.