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Well, I'm Matt Pelton, and I'm very glad to have the chance to talk to you today about some work we've been doing over the last few years in coupling together single quantum dots with plasmonic-metal nanostructures, and, in particular, reaching the strong coupling regime between single emitters and plasmonic nanostructures. And when I talk about the strong coupling regime, I mean this in the context of cavity quantum electrodynamics.

So since the '80s, it's been recognized that you can modify the properties of emitters by placing them inside optical cavities. So the first experiments that were done were to modify the spontaneous-emission properties of emitters by putting them in optical cavities, basically between a pair of mirrors, because spontaneous emission really isn't a property of an emitter by itself, but as a result of the interaction between that emitter and its surrounding electromagnetic compartment.

So if you change that environment by placing the emitter inside an optical cavity, we're changing the density of optical states available for the emitter to radiate into. So you can enhance the emission rate of the emitter into those cavity modes. So that's what's become known as the weak-coupling cavity QED regime, the so-called Purcell enhancement effect.

But if you can make that coupling between the emitter and the cavity photons large enough, then you can get into the so-called strong-coupling cavity QED regime, where instead of the light from the cavity being irreversibly lost to the environment, it will be reabsorbed by the emitter. You'll have coherent energy exchange back and forth between the excited state in the emitter and the cavity photons before energy is lost from the system.

So that oscillation of energy back and forth, what's known as the vacuum Rabi oscillations, corresponds in the frequency domain to a splitting in the spontaneous-emission spectrum of the emitter inside the cavity, with the splitting determined by the coupling strength between the emitter and the cavity.

So if you're thinking in terms of energy state, instead of having the isolated energy space of the emitter and the photon inside the cavity as the energy eigenstates miss this step, now you form new hybrid states, new normal modes, that are a combination of emitter and the photon, so a so-called polariton state.

And the interest in forming these polariton states is because of the possibility to exploit those in quantum information applications. It has been shown that you can use this strong coupling to effectively induce nonlinearities at the single-photon level when light interacts with this coupled system, which means that you can have photon-photon interactions at the single-photon level.

You can have information transfer between the state of a photon and the state of an emitter, say, the spin state of an emitter inside the cavity. You can entangle that spin state with a polarization state of the photon. You can use this as a way of transducing optical information from the propagating photon to a solid-state system.

So within that context, that we're interested in looking at plasmonic-metal nanostructures that are interacting with some single emitters, because we can think of plasmonic-metal nanostructures as optical cavities, but the ones that are not limited by the diffraction limits.

So instead of confining light by reflection, plasmonic nanostructures confine optical fields by coupling them to material excitations to collective oscillations of electrons inside the plasmonic-metal nanostructure. And in that way, the optical fields can be confined to really nanometer-scale volumes within and near the metal nanostructures.

With that ability to confine light to localize it on the nanometer scale, this motivated a lot of the interest in the applications that plasmonics have to offer, from imaging to data storage to sensing, and in our case, strong light-matter interactions at the quantum level.

And this ability to modify the properties of emitters by coupling them to plasmonic modes isn't something new. It's actually something that goes back to the early days of plasmonics. I'd say that modern plasmonics started with the discovery of surface-enhanced Raman scattering, the fact that you can get Raman signals from molecules adsorbed onto roughened metal surfaces or in aggregates of metal nanoparticles, and for orders of magnitude higher than you would get from the molecules alone.

And it was recognized shortly after the first experiments in the '70s that the main reason for this massive enhancement of Raman scattering signals was the localization of sub-nanometer-- or sub-diffraction-- limited confinement of light around the metal nanoparticles. So if you can enhance from a scattering by confining light like that, you can enhance any optical process, including spontaneous emission.

And in our context, we can see that as a form of recoupling cavity QED, where the plasmonic-metal nanostructure is serving as a cavity. And then you can get some enhancement factor that's proportional to the quality factor of the plasmonic or photonic cavity and inversely proportional to the mode volume. So the longer the photon lifetime is, or the smaller the volume otherwise confined into, the stronger the interaction you have between light and the emitter.

And plasmonic-metal nanostructures are in a way very lousy resonators. They have low quality factors of the order of 10 to 100. On the other hand, since they're not limited by diffraction, they can have extremely small mode volumes. And this can more than make up for the loss of quality factor that you have in counting to the material expectations and get Purcell factors that are larger than possible with any photonic counting.

So this ability to get very high Purcell factors using plasmonic-metal nanostructures naturally leads to the question of whether we can reach the strong coupling regime with the same kind of plasmonic structures.

And since we are seeing this in the cavity QED regime, we can use the same language, the same mathematics, the same models to describe plasmon-exciton coupling as we use in cavity QEDing, particularly the so-called Jaynes-Cummings-Hamiltonian, the sort of canonical model Hamiltonian where we have a two-level system interacting with a set of bosonic modes, in this case representing the plasmons inside the metal nanostructure wrapped in bare photons inside the cavity and an interaction that just simply exchanges information-- I'm sorry-- into energy between the plasmon and the two-level system.

And this leads to a mode splitting that's given by the coupling strength, but this doesn't take into account any damping inside the system. And, of course, the damping in this plasmonic discipline was especially strong, so if we want to have a realistic model of its-- we need to include that damping.

For mechanically, this is done using the Heisenberg-Langevin equations or other equivalent quantum models, putting in these Lindblad operators to describe the damping of the system. And this whole set of equations can be solved numerically, you know, in the time domain, in the frequency domain, and to understand the spectral properties of this complex system.

But it turns out that in the regime that we're interested in, particularly for the strong dephasing of plasmons and room-temperature emitters, so strongly dephased emitters as well, coherences in the optical field are not important. And you can treat the optical fields classically, so as complicated equations reduced to semiclassical or Maxwell-Bloch equations. And these Maxwell-Bloch equations can be further simplified if you're interested only in the linear optical response of the system.

So in the regime of weak excitation of the system, where the emitter stays primarily in this ground state, you can describe using purely classical equations that are equivalent to just a pair of coupled classical harmonic oscillators, with the dipole moment of each oscillator taking the place of the displacement, a coupling strength that represents this near-field dipole-dipole coupling between the plasmon and the emitter, and then the driving term that represents the action of the external electromagnetic field on the plasmon.

And since the plasmon in the metal nanoparticles has an optical cross-section that's much larger than the cross-section of the single emitter, we consider direct driving only of the plasmonic-metal nanoparticle. And then the emitter is driven only through its coupling to the plasmon. And this set of couple of equations says it can be solved analytically. And we can see what happens as we put in realistic parameters for a plasmon, for a single quantum dot, and just increase the coupling strengths between the two of them.

So for weak coupling strengths, we have the weak coupling regime, the Purcell enhancement regime, where the lifetime of the emitter is reduced, but it's for the luminescent spectrum. The scattering spectrum from the plasmonic-metal nanostructure are basically not changed from what they would be if it weren't coupled to each other.

And if we go to coupling strengths that are much larger, both of-- from the luminescent spectrum and the scattering spectrum split into two distinct peaks separated by the coupling strengths that represent these new polaritonic modes, the hybrid plasmon-exciton mode to the system.

But in between, we have, like all the intermediate coupling regimes, or the high-cooperativity regime, where you don't have splitting in the spectrum of the quantum dot photoluminescence, but you have this clear dip in the scattering spectrum from the plasmonic-metal nanoparticle.

So the two peaks here are not represented in normal mode splitting, but are actually representing destructive interference between the quantum-dot plasmon or the emitter-- sorry-- the quantum-dot dipole moment or the emitter dipole moment and the plasmon dipole moment that are inducing a transparency, as we call it, exciton-induced transparency in the spectrum of the metal nanoparticle.

So we really have two distinct regimes for this high cooperativity or intermediate coupling regime and depend on the ratio of the coupling strengths to the damping rates from the plasmon, and the [INAUDIBLE] is a great emitter, or the strong coupling regime which depends on simply the ratio of the coupling strength to the sum of the γ rates inside the system.

In either case, what we need is a coupling strength that's large compared to the rate at which energy is lost or dephasing occurs inside the system. And a photonic cavity is-- there's coupling strengths that's strictly limited by the diffraction limit. Since the mode volume is limited to be at most-- or at least half a wavelength on a side, the coupling strengths has a physically imposed limit on it because of this diffraction limit, which means that the only way to reach strong coupling in a photonic cavity is to have sufficiently narrow cavity in linewidths and emitter linewidths.

The cavity in linewidths is usually taken care of because photonic cavities, such as photonic crystal cavities, can have nearly diffraction-limited mode volumes while having very high quality factors, on the order of 10 to the fifth or even higher. On the other hand, emitter linewidths, at least in solid-state systems, are going to be firmly broadened. And so the only way that you can reach the strong coupling regime is either to have an isolated system, like a single atom, or to take your solid-state system and cool it down to cryogenic temperatures.

On the other hand, for plasmonic modes, plasmonic cavities, the mode volume is no longer limited by diffraction, so you can have much larger coupling strengths. On the other hand, the plasmon linewidths is much larger, and plasmon lifetimes are inherently on the order of 10 to 100 femtoseconds at most, which means that you have to reach even stronger coupling strengths, very small mode volumes, in order to reach the strong coupling regime.

But on the other hand, once you've reached that, you are well over the thermal linewidth, and so you can reach strong coupling at room temperature in these systems if you could make your mode volume small enough.

And the way to make the mode volume small enough for it to use the so-called gap plasmons-- so again, it's the mode for a long time, but you can confine light with a pair of metal nanoparticles much more tightly than you can confine light with a single metal nanoparticle by itself. When you bring two metal nanoparticles close together, the electron oscillations and plasmons interact with each other electrostatically, confining light into the gap between the two particles.

The same sort of thing happens if you take a metal nanoparticle-- put it above a metal surface. You can think of the dipole of the plasmon in this particle interacting with its image dipole inside the metal film and, again, confining light in the gap between the particle and the film.

So if you then place an emitter into this gap, and you make the gap small enough so that it really confines light on a nanometer-scale volume, we should be able to reach those very small mode volumes and the very high coupling strengths that will enable cavity QED at room temperature with a single emitter.

And the single emitters that we choose are colloidal quantum dots. We choose these over molecules because they have larger transition dipole moments, and since the coupling strength is proportional to the dipole moment, that gives us another boost in trying to reach strong coupling.

pling. They're also relatively photostable compared to molecules that are less subject to photobleaching or to degradation when you shine light on them.

But we continue the transition frequency of these quantum dots by changing their size, so then we can match the quantum-dot transition to the resonance frequency of this plasmonic mode. And compared to, say, epitaxial structures, like indium arsenide, gallium arsenide, surface-grown quantum dots, these are not embedded in some larger semiconductor structures. So they're relatively easily integrated into this plasmonic structure.

And the way we did that in our first experiment, and sort of randomly redecorated the surface of quasi-spherical gold particles with a small number of quantum dots and put them down on the metal surface. So this was done in collaboration with the group of Marie-Christine Daniel in the Chemistry Department at UMBC, where they coated the surfaces of quantum dots and the surfaces of the metal particles with complementary molecules, linked them together at concentrations so that only a few particles would bind to each metal nanoparticle, and then just deposited these metal nanoparticles, decorated with quantum dots onto a metal surface.

And most of the time, we don't get strong coupling, but every now and then, the quantum dot will land at just the right place between the gold particle and the silver film to give a strong couple.

So to show the data for that, again, like I said, most of time we get weak coupling, so if you look at the decay of the quantum dot, it's much faster than it is by itself. We have a strong Purcell effect with large enhancement of the decay rate, but if you look at this green photoluminescent spectrum, the quantum dot, or the blue scattering spectrum from the plasmonic-metal nanostructure, they're both basically the same as they would be for the particles by themselves.

But if you have a patient-enough graduate student who's willing to look at a few hundred particles, eventually now and then you'll find one that has a quantum dot in the right place and reaches either the intermediate or strong coupling regimes. So sometimes you have this clear dip and a scattering spectrum that lines up with a maximum in the photoluminescent spectrum and represents this exciton-induced transparency or a destructive interference between the dipoles of the plasmon and the exciton.

And sometimes you're really lucky, and you get splitting of both the plasmon scattering and the photoluminescence from the quantum dots, or really both of these are coming from the hybrid states of plasmon and exciton in this strongly coupled Rabi-split regime.

So you can see here, there's a lot of heterogeneity and a lot of variation from one particle to another in terms of what that coupling strength is. So instead of just trying to understand the origins of that heterogeneity, we turn to finite-element calculations, where we took a gold nanosphere quantum dot, and that replaced the quantum dot a little bit off to the side or right in between the gap between the two of them.

And we can see we clearly have weak coupling in the scattering spectrum here, but really only reaching intermediate coupling. Even in this case for the quantum dot, it's exactly where we want it to be. And I think the reason for the discrepancy between this calculation and what we see experimentally is, experimentally, we don't have these nice, round, spherical ball particles. We have these faceted quasi-spherical particles, and the quantum dots can be on one of those facets or at the vertex where those facets meet.

And so if we sort of do a cartoon representation of this facet being in our calculations, we can get stronger and stronger coupling, depending whether we put

them edge or on the corner, that the field becomes more and more tightly confined. So this is really showing that detailed control over the nanometer-scale structure of our plasmonic nanoparticles or plasmonic-metal nanostructure is necessary to reliably get strong coupling at the single-particle level.

So towards getting that coupling deterministically and reliably, we collaborated with the group of Markus Raschke at the University of Colorado, replacing that gold particle with a scanning probe tip that could be placed exactly on top of the quantum dot and get reproducible strong coupling with the single quantum dots, again, from moving from one quantum dot to another over and over again. I have a separate talk on this at this digital forum, so I will only mention this here and invite you to watch that talk as well, if you're interested.

And to go a little bit more into the details of how we did this or how we analyzed this data, while other time, people will look at simply the peak separation in a photoluminescent spectrum or in a scattering spectrum and take that to be the coupling strengths and use that to determine whether they're in the strong coupling or weak-coupling regime.

So we've seen this is especially problematic if you're looking at the scanning spectrum because the exciton-induced transparency will give you two maxima, even when you're clearly not in the strong-coupling regime. If you really want to know the coupling strengths, you need to do a full lineshape analysis.

And what we did is we went back to the published literature and to our own data and fit the photoluminescence spectra or the scattering spectra to the predictions of this coupled oscillator model and seeing whether we're in the strong coupling regime, where this number should be larger than 1, or in the exciton-induced transparency, where this cooperativity should be larger than 1.

And you see that the threshold for strong coupling is not a strict one. It's more of a rule of thumb than a real threshold. So most of the results are very close to being at the edge between strong coupling and weak coupling, with one previous result using a scanning probe technique that had a more complicated model for what was going on, according to that model was thought in the strong coupling regime.

On the other hand, most of these are not in the high-cooperativity regime, so the applications that would exploit cooperativity are not accessible here, except for this one previous report, whereas in our cases, we were able to reach both the strong coupling regime and the high-cooperativity regime with a reasonable margin of safety.

And so like I said, this strong coupling has been largely investigated for-- on information applications, but I think maybe a nearer term application for these room-temperature systems would be classical information processing because its high cooperativity or exciton-induced transparency opens up the road for large nonlinearities in this coupled system.

This, for example, is a calculation we did some time ago, where we put a single quantum dot in the computer between a pair of metal nanoparticles and looked at its absorption spectrum as we increased the intensity of light on this coupled system. You can see as the intensity increases, this transparency dip goes away, and the full scattering of the metal nanoparticle is recovered.

And this transition between being weakly absorbing or strongly absorbing at the location of this dip occurs at moderate optical intensities. But that means since the footprint of this thing is so small, since it's really a nanoscale structure, the total power required to switch from absorbing to not absorbing would be very small.

And the reason for this nonlinearity is simply the saturation of the quantum-dot transition. So once the quantum dot is in this excited state, it's not going to absorb more light as transition dipole moment goes to zero. It's as if it's not there, as far as the metal nanoparticle is concerned. There's no more interference, and the metal nanoparticle's scattering or absorption is fully recovered.

Since this happens at very low optical powers, since it can be ultrafast because the recovery time of the system would be on the picosecond timescale, and since the whole thing is only about 10 nanometers on the side because of really an ultrasmall device, too, we think that it could serve as the critical element in future all-optical information processing as an ultrafast and ultrasmall, ultralow-power optical modulator.

They could potentially be used in more exotic or optical architectures, such as all-optical neural networks, where it would serve as a nonlinear activation function in the node or neuron of a neural network. So the job of doing a weighted sum to be handled interferometrically with phase-diffusing combiners, but you need some small neural--nonlinear optical element to give the activation function inside this computer.

And they showed, in principle, that the nonlinearity provided by these coupled quantum-dot metal nanostructures would enable all-optical neuromorphic computing at the speed of light, meaning that the computational time would simply be limited by the propagation time of light from one end of the neural network to the other.

Of course, if you want to make devices out of these things, we need to make them reliably. The 1% or 2% device yields we had before is not going to do it. And using a scanning probe is great for experiments and for quantitative understanding of what's going on, but that's not a way to make devices either.

So what we're doing right now, again with the collaboration of the group from Marie-Christine Daniel, is to modify that self-assembly technique where we're attaching single quantum dots to spheres, now to use anisotropic particles, like gold nanorods, gold bipyramids, where we can selectively attach single quantum dots to the ends of these particles, and then link them up in the chain, a gold particle, quantum dot in the gold particle, and reproducibly get strong coupling in a macroscopic number of these things. And we can synthesize in the simple tin solution.

So with that, I'd like to wrap up. Thank you for watching my presentation and invite you to email me any questions that you have, just saying that we can meet in person in San Diego. I would be very happy to hear from you by email if there's anything that you would like to ask.