Ultrafast Optics and Spectroscopy
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Lecture – 30
Ultrafast Physical Chemistry: Solid State

Welcome to module 12 of the course Ultrafast Optics and Spectroscopy. In this module we will go for different ultrafast processes involved in solid, semiconductors, nanoparticles and quantum dots, we will go over the content very briefly.

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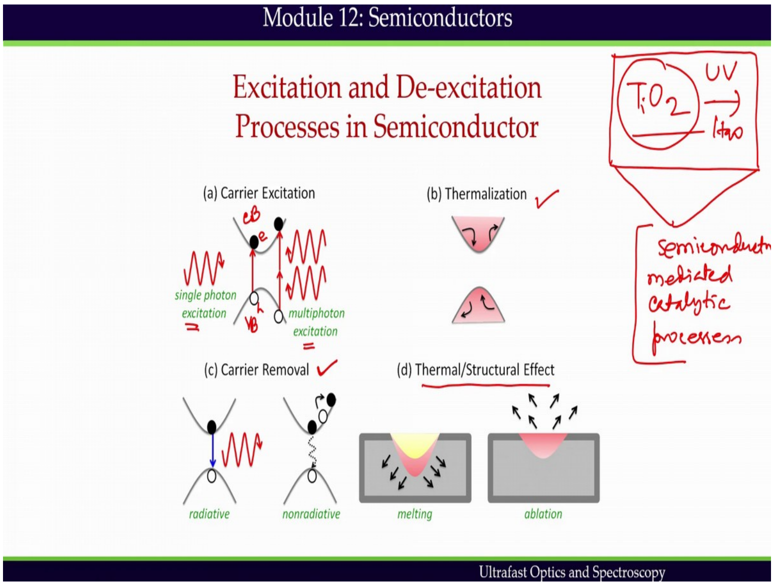

Module 12: Semiconductors

Excitation and De-excitation Processes in Semiconductor

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Titanium dioxide TiO 2 is a semiconductor and titanium dioxide, when it was irradiated with ultraviolet radiation it was found that this semiconductor in fact, metal oxides are semi-conductor. Metals are conductors but, metal oxides are semi-conductors, UV excitation of TiO 2 can dissociate water or split water with the help of UV radiation. So, it can dissociate water with the help of UV radiation and this discovery, that photoexcitation of TiO 2 can split water drew our attention because, we realise that semi-conductors can be used in the field of heterogeneous photocatalysis.

Photocatalysis is a catalytic conversion process induced by photon, upon optical excitation a semi-conductor undergoes several stages of relaxation before returning to the initial state. And because of this discovery we took interest in semi-conductor mediated catalytic processes; semi-conductor mediated catalytic processes. And because of that we were interested in looking at what are the ultrafast processes involved after photoexcitation of a semiconductor. Optical excitation energy is first transferred to the electrons and then to the lattice of the semiconductor.

From a simplicity point of view all electrons in semiconductor can be considered to be part of an electron reservoir or heat bath. And all iron cores can be collectively consider to be a part of a phonon bath or lattice heat bath as heat capacity of electron path is lower than that of lattice bath, optical excitation energy are first transferred to the electrons if photon energy matches the band gap.

So, let us say this is the band gap, the direct band gap, this is conduction band, this is valance band and this is the valance band and this is conduction band. And if we excite the electron from valance band to the conduction band first, we can do it with the help of single photon excitation or multiphoton excitation.

But, following this electronic excitation we can have many processes opened up; the first one is going to be thermalization, then carrier removal or recombination of electron whole pair. When we create, when we excite the electron from valance band to the conduction band, we create a hole in valance band and an electron in the conduction band.

And this electron hole pair can recombined in the carrier removal step and finals process which can be opened up after carrier removal is the thermal or structural effect. We will go over all these effects systematically; the first step is carrier excitation. What happens in 100, 10 to 100 femtosecond after the carrier excitation?

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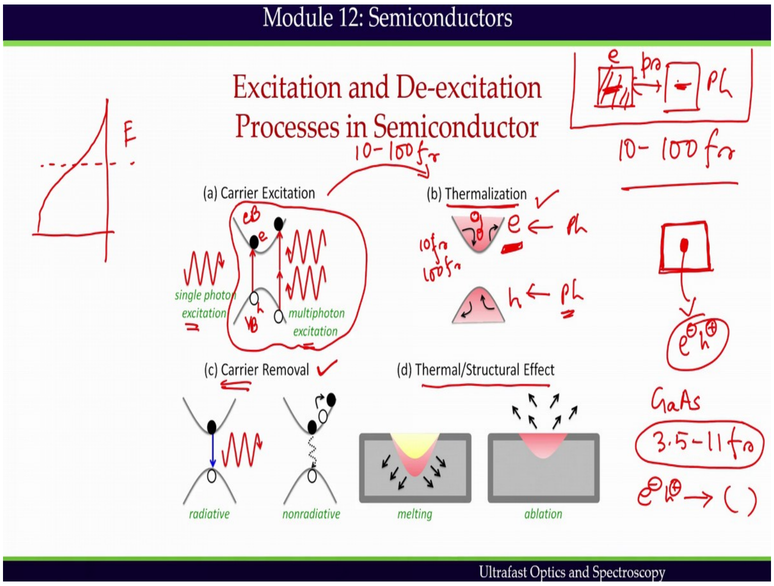

Module 12: Semiconductors

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So, we are looking at the time of 10 to 100 femtosecond first, 10 to 100 femtosecond time. When the excitation photon is larger than the band gap, single photon absorption occurs and electrons from the valence band are excited to the conduction band, multiphoton absorption can also occur when the direct band gap is greater than the photon energy. Whenever an electron acquires energy sufficient to move from valence band to the conduction band, a free hole is created in the valence band and an excited electron resides in the conduction band.

However, an electron and a hole attract each other. They will so, here whatever picture we are showing the excitation picture, this is in terms of the energy profile diagram. But, if you think about the material, where the excitation is going on in this box size we will create electron and hole. Consequently, electron hole pairs are general generated due to optical excitation in semiconductor.

This kind of electron hole pairs is generated. The electromagnetic field which causes electron hole pair formation can also stimulate them to recombine which is called stimulated emission that is also possible. However, the coherent affect oscillatory formation of an annihilation of electron hole pairs are only observed on the time scale of 10 femtosecond.

For gallium arsenide this is semiconductor or this semiconductor, the coherent time scale is found to be 3.5 to 11 femtosecond, within this time scale we can create electron hole pair and then we can annihilate also. So, creation and annihilation of electron hole pair is an oscillatory process and that can occur within this 10 femtosecond time scale. Once the oscillatory coherence effect is lost within 10 femtosecond time scale the carriers, here when you say carriers it is actually electron hole pairs.

When electron and hole pair are relaxing freely, then we have to remember that electron we will relax in the conduction band and hole we will relax in the valence band. And, when its relaxing it means that this whole relaxation process is called thermalization. Thermalization does not mean that it's thermalizing or in it is becoming in equilibrium with the whole body.

It is reaching equilibrium, a local equilibrium within the spot of the excitation. Immediately after optical excitation the carrier distribution does not follow a Fermi Dirac distribution. Generally, Fermi Dirac distribution how does it look like? This is the probability, this is the energy and this is kind of a Fermi energy and distribution look like this, this is the hot distribution.

Now, immediately after optical excitation the carrier distribution does not follow a Fermi Dirac distribution, that is thermalized distribution of the Fermi free electrons which comes from Fermi free electron gas model pointing to a highly not thermal distribution. So, immediately following excitation we have very non-thermal excitation that will thermalize, within the spot size of the excitation.

It takes 10s to 100s of femtosecond for the non-thermal carrier distribution to approach a Fermi Dirac distribution. So, this process occurs 10 to 100 femtosecond time scale, within this time scale we achieve some kind of thermal distribution. So, we will be able to define a temperature for the electrons and the holes. Carrier-carrier scattering is a two body inelastic scattering process in which total energy in the excited carrier system remains unchanged.

And through which a hot Fermi Dirac distribution of the carrier is achieved, in semiconductor another inelastic scattering is initiated concurrently during the first several 100 femtoseconds after carrier excitation which is called carrier phonon scattering. So, this electrons and holes they can also scatter with the phonon, that is the lattice core and they can exchange energy. We have discussed all electrons can be considered to be an electron heat bath and all phonon can be considered as phonon heat bath and they can exchange energy.

In carrier phonon scattering process free carriers lose energy by a phonon excitation, phonons will be excited. Phonons are spontaneously de-excited transforming energy to the lattice, relaxation of carrier through carrier phonon scattering process; however, features several picosecond time scale. So, we have to remember carrier-carrier scattering and carrier phonon scattering has a big time difference.

If it is carrier-carrier scattering which means high energy electrons is scatting with low energy electron that is carrier-carrier scattering; that will occur 10 to 100 femtosecond, 10 femtosecond to 100 femtosecond. But, if it is electron or carrier phonon scattering that will occur in picosecond time scale, there is a time scale difference between these two processes.

In the end of the several picoseconds the hot carriers and the lattice reach thermal equilibrium. In the end within the spot size the first equilibration we get is in the carrier first, then we will be able to define a Fermi Dirac distribution for this carrier. Then within picosecond time scale we will reach an equilibrium between electron or the carrier and the phonon within the spot size. Third step is carrier removal; so, once we have reached this equilibrium, local equilibrium between carrier and phonon, then the third step is going to be the electron hole pair recombination.

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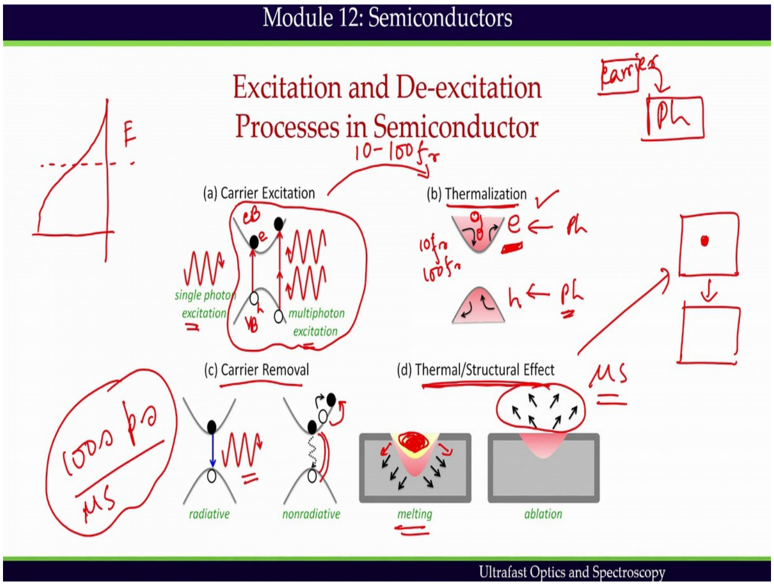

Module 12: Semiconductors

Excitation and De-excitation Processes in Semiconductor

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Electron hole pair recombination it means carrier removal and this is the third step, where we remove the carrier. In several picosecond time scale and the time scale is going to be 100s of picoseconds, excited electrons relax back to the bottom of the conduction band and hole relaxes back to the top of the valence band. Carriers are then removed by recombination of electrons and hole both radiative and non-radiative processes can play roles in carrier annihilation. During radiative recombination the carrier energy is given up in the form of a photon and that gives luminescence which is shown here.

Non-radiative recombination processes may occur through Auger process or defect recombination which means when this electron hole pair recombining, I can have another carrier excited to the electronically excited state; that kind of process can happen. This is non-radiative process which means that the energy which is released by this recombination can be used some other excitation that is non-radiative process.

During Auger recombination an electron and a hole recombine and as a result excess energy excise and electron higher in conduction band. In defect of surface recombination on the other hand, the excess energy is given to a defect or surface state. In semiconductor material the time scale of carrier removal that is electron hole pair recombination ranges from several 100 picosecond to several 100 microseconds. So, this can be this can extend up to even microsecond times scale.

It depends on how quickly it is going on or different system. The final step is the thermal and structural effect. The moment we have all this recombination is over, then we observe these thermal or structural effect and that occurs in micro second time scale. We have mentioned that the lattice temperature gradually increases, the phonon temperature remember carrier finally, will give the energies to phonon.

And finally, phonon will be excited, the lattice will be excited and if the lattice is excited then I can have thermal diffusion process which can cool down the hot lattice of the photoexcited regime. So, let us say this regime is photo excited and I can have a cooling down the heat diffusion which can occur. This cooling can eventually send the photo excited regime back to the ambient value of the time scale of microsecond.

In micro second time scale wherever we have done the photoexcitation that photoexcitation regime can go back to its original or its earlier condition, due to this thermal structural effect cooling effect. However, instead of thermal diffusion if the lattice temperature eventually exceeds the melting point temperature then the region of the photoexcited region can be melting. Or, it can change the phase and transform to superheated solid until the photoexcited region nucleates.

The liquid or gas phase may expand into the material starting from sides at the surface. Thus, the energy deposited by the laser falls may be converted to kinetic energy of the lattice ions; with this kinetic energy, ions, atoms, molecules or clusters can also leave the surface. It can actually undergo ablation is a non-thermal evaporation. Following ablation re-solidification can also cool the lattice in photoexited regime.

However, in this case material may not necessarily revert back to its original structure or phase following melting or abolition of photoexcited regime. Here although we have discussed four different steps, we should keep it in mind that these channels may not appear sequentially. One step may substantially overlap with one another and that is why times of spectroscopy can distinguish different steps, if we would like to explore the processes individual processes.

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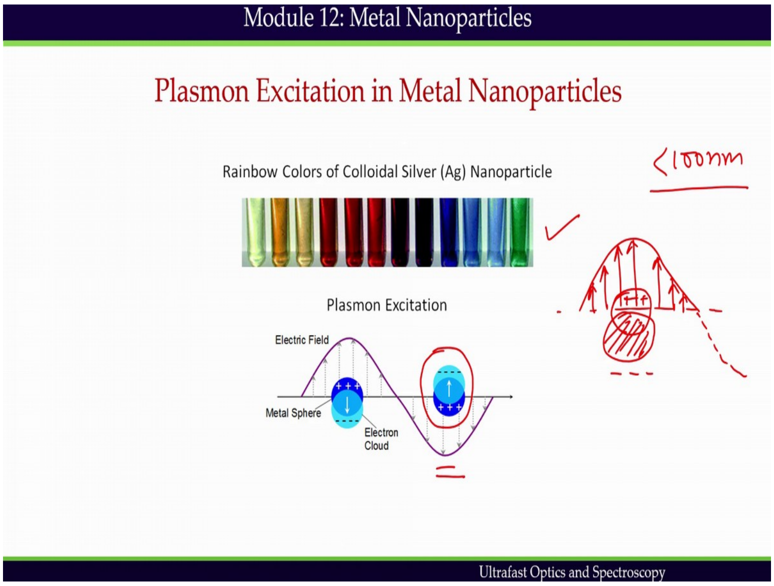

Module 12: Metal Nanoparticles

Plasmon Excitation in Metal Nanoparticles

Rainbow Colors of Colloidal Silver (Ag) Nanoparticle

Plasmon Excitation

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Now, we will take a look at the metal nanoparticles, metal nanoparticles are metal particles with typical dimensions less than 100 nanometer. So, any metal particles which are less than 100 nanometer we call them metal nanoparticle. When solutions of metal nanoparticles are illuminated by white light, they exhibit different colours. An illustrative examples of silver nanoparticle is shown here, different colours can be seen for different sizes and shape of the nanoparticle.

The intense colour of nanoparticle solution is determined by the collective excitation of all electrons in the nanoparticles which is called plasmon excitation. Shown here I have nanoparticle and the moment I have electromagnetic radiation interacting with it, electromagnetic radiation is changing its field direction in every half cycle. So, in this cycle what is going on? I have field directed towards this that is why electron density will feel a force along this way.

Field is nothing, but force perimeter charge that is why electrons will be polarized and my ion cores are polarized. So, this polarization positive negative charge separation occurs within this half cycle. In the next half cycle field has changed and that is why polarization has changed which means that the whole electron density will oscillate as the field of the electromagnetic radiation is changing over the optical cycle.

This oscillation is called plasmon excitation, plasmonic excitation which determines the optical properties on metal nanoparticles is technologically very important phenomenon. Because, it finds numerous applications including phototherapy of cancer using gold nanoparticles. In phototherapy plasmons of gold nanoparticles are excited with light.

This photoexcitation ultimately results in release of heat energy which kills the targeted cancer cells. Therefore, it is of both fundamental and a practical importance to understand the excitation and relaxation processes of plasmons in metal nanoparticles. The first step is plasmon excitation, in the photo excitation of nanoparticle system. From a simplistic point of view all electrons in the metal nanoparticles can be described collectively as plasma.

In metal nanoparticles with size is smaller than the wavelength an optical penetration depth of the excitation pulse, excitation light plasma can be collectively excited as shown here. The collective excitation of the plasma in a metal nanoparticle is called plasmon. Due to interaction of metal nanoparticles with the electromagnetic wave electron gas gets polarized and restoring force is created by the ion core which results in this plasmonic oscillation. The collective oscillation can be integrated as a displacement of the centre of mass of all electrons in the particle against the positively charged background of the ion core. Thus, metallic nanoparticles act as oscillators and the corresponding resonance behaviour determines the optical properties of the nanoparticles.

The resonance frequency of the oscillation is determined by the dielectric properties of the metal and surrounding medium and by the size and shape of the particle. The collective charge oscillation causes a large resonant enhancement of the local field as well inside and near the particle.

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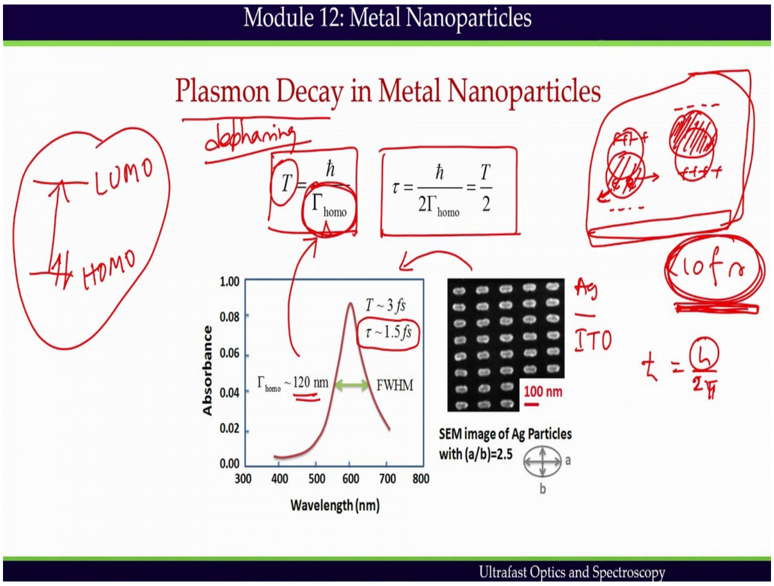

Module 12: Metal Nanoparticles

Plasmon Decay in Metal Nanoparticles

$$T = \frac{\hbar}{\Gamma_{\text{homo}}}$$

$$\tau = \frac{\hbar}{2\Gamma_{\text{homo}}} = \frac{T}{2}$$

SEM image of Ag Particles with (a/b)=2.5

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So, these are the different features of the plasmonic excitations. The second step of plasmon decay is once we excite that, once we have created that oscillation this polarization negative, positive. And then in another cycle we have positive, positive; once we have turn on this oscillation the next step is going to be plasmon decay. This occurs in few 10s of femtosecond. So, it could be of the order of less than 10 femtosecond time scale this decays, this plasmatic excitation.

Here we remind ourself that this plasmonic excitation is not like a HOMO LUMO transition. Generally, in photochemistry we discussed the HOMO LUMO transition; this is Highest Occupied Molecular Orbital, this is Lowest Unoccupied Molecular Orbital. And we excite electron because this is fully occupied, doubly occupied and this is empty that is why we excite an electron to the excited state and we undergo and photoexcitation.

We have to remember here the plasmonic excitation is not like a HOMO LUMO translation, its just like an oscillation induced by the electromagnetic radiation. No electron during this oscillation, no electron has been excited to the empty excited state; it is collective oscillation of the electron induced by the field of the electromagnetic radiation.

A plasmon is formed when the whole electron density of a metallic nanoparticle oscillates due to influence of an external electromagnetic field. Following plasmon excitation; however, this coherent motion of the electrons is easily destroyed by for example, different scattering events which annihilate the phase coherence of the coherent oscillation. So, what might happen during this oscillation it may so, happen that I have an electron which is trying to go this way and another electron is trying to go this way.

And this mismatching phase will destroy ultimately destroy this coherent oscillation of all electrons. One can imagine this destruction of phase coherence or dephasing of the plasmon by simply kicking an electron out of the coherent oscillation with the help of impurity or another electron. So, the very first step of plasmon decay is dephasing, this is called dephasing; dephasing these electrons are not coherently oscillating.

They are trying to diphase, the dephasing time this T capital T corresponds to the time scale on which the decay of the coherent plasmon electron oscillation takes place. And the electron oscillation preserves the memory of the optical phase of the excitation pulse. The dephasing time can be estimated by ultraforce spectroscopy as well as with the help of absorption spectroscopy of the metal nanoparticles.

The dephasing time; the dephasing time is related to homogenise line width, that is capital tau[τ] shown here by capital tau. This is the line width, there is the full width half max of the absorption spectrum. This is the natural line width related to uncertainty principle if. What does it mean? It means that if we observe the absorption spectrum as shown here, the full width half max of the absorption spectrum can give me the line width with the help of the it can give me the line width with first.

And, this line width will give me the dephasing time, the lifetime of the plasmonic excitation. Here T is expressed in second capital tau homogenous line width is expressed in eV and h is Planck constant although, this h by 2 pi and this h is Planck constant. The plasmon lifetime is then expressed by tau which is nothing, but T by 2. Typical dephasing timescale of the my plasmon determined by both the line width and time resolve measurements on metal ion particles is found to be of the rock or of the order of 10 femtosecond.

So, within 10 femtosecond it will decay and because it is 10 femtosecond lifetime the width would be very very long because, of the uncertainty principle delta t delta E is nothing, but by 2 something like that. So, in it depends on the uncertainty principle. The experimental determination of phasmon dephasing time scale of nanoparticles is pretty daunting task, not because it is it occurs in very short time scale, but because of some other reason. The reason is that any synthetic procedure to prepare nanoparticle, any synthetic procedure renders nanoparticles with broad distribution in particle size and shape.

And, as I mentioned before that the absorption spectrum or in other words the diffusing time will strongly depend on the size and the shape of the nanoparticle. So, if you are synthesizing an nanoparticle system having very broad distribution of particle size and shape, then what will happen this randomness in size and shape results in a strong inhomogeneous broadening of the optical absorption spectrum. Different particle exhibit slightly different absorption spectrum and that is why finally, we get a broad spectrum.

Since, the magnitude of the inhomogeneous broadening is unknown is not a known quantity, for any system the homogeneous line width that there is we are interested in this homogeneous line width that cannot be extracted; if the particles exhibits broad size and shape distributions. That is why starting nanoparticles is a problem with regard to the absorption spectroscopy as well as time resolve spectroscopy, because of the broad distribution. And each particles has its own behaviour finally, we see an average behaviour of the particles.

But, to understand size dependent and shape dependent behaviour we need to have a synthetic procedure which will systematically produce very identical nanoparticles. One potential way to circumvent the above inhomogeneous broadening problem is by analysing a lithographically produced regular two dimensional array of identical metal nanoparticles. Lithography is a technique by which we can create very identical nanoparticles. Advance electron beam lithography technique allows want to position metallic nanoparticles of any shape, size and orientation in any arrangement pattern on the flat substrate.

Here an example is given, this is the silver nanoparticles, these are silver nanoparticles and this array has been prepared on indium tin oxide flat surface. These silver nanoparticles were fabricated by means of electron beam lithography and the absorption spectrum which has been recorded from this silver nanoparticles is shown here. From the experimentally obtained line width, this line width was 120 nanometer and this 120 nanometer line width, if we convert it to its dephasing time then we get 1.5 time 1.5 femtosecond as our time scale of plasmonic decay. So, this suggest that the plasmon excitation and its decay can be very very fast.

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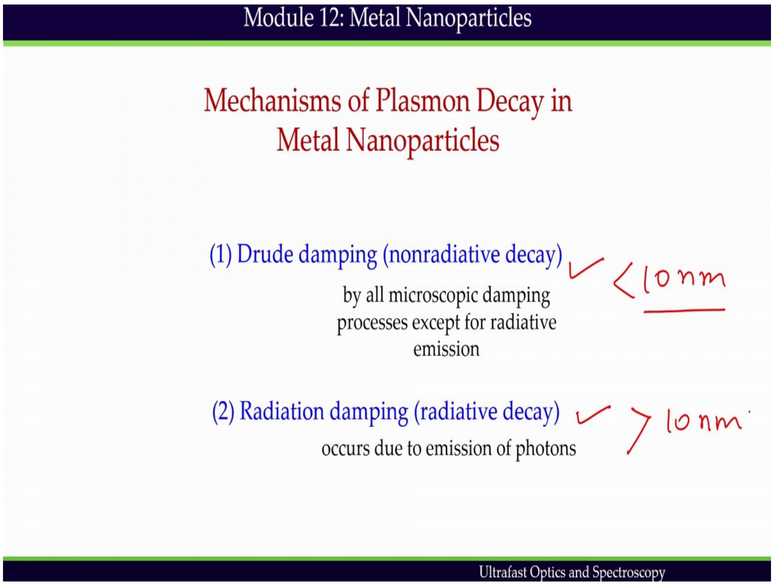
Module 12: Metal Nanoparticles

Mechanisms of Plasmon Decay in Metal Nanoparticles

(1) Drude damping (nonradiative decay) ✓ $< 10\,\mathrm{nm}$

by all microscopic damping processes except for radiative emission

(2) Radiation damping (radiative decay) ✓ $> 10\,\mathrm{nm}$

occurs due to emission of photons

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But, what is the mechanism of dephasing, the plasmonic dephasing? Depending on the size, size distribution, shape, dielectric constant of the surrounding medium two potential mechanisms for dephasings are considered. The first mechanism is Drude damping mechanism which is nonradiative decay and another one is radiation damping, that involves radiation, emission.

Drude damping involves dephasing by all microscopic damping processes except for radiative emission. Drude damping can occur due to decay of the fixed phase correlation between the individual electronic excitation of the whole oscillator ensemble. This type of damping is called Drude dephasing because, it does not involve radiation or emission and can be caused by local inhomogeneity of the nanoparticles. Drude damping can also occur due to a transfer of energy into quasi particles by formation of electron hole pair by surface or electron-electron scattering.

The radiation damping on the other hand occurs due to emission of photons which results in luminescence. In general, Drude damping is dominant for metal nanoparticles which size is below 10 nanometers. So, if the particle size is 10 nanometre generally it is found that they are the plasmon would decay through Drude damping. But, the particle size if it is larger than 10 nanometre, bigger particles we get radiative emission.

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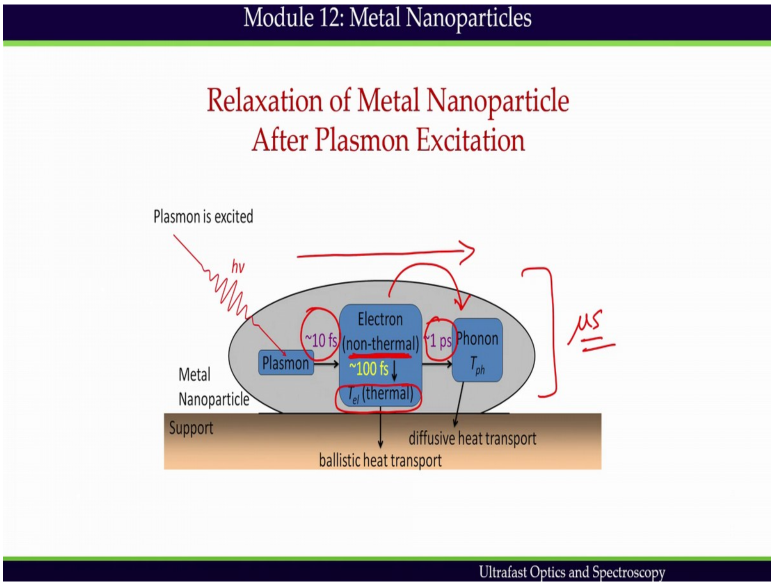

Module 12: Metal Nanoparticles

Relaxation of Metal Nanoparticle After Plasmon Excitation

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As pointed out in the previous slide radiation damping results in luminescence; however, due to Drude damping finally, quasi particles electron hole pair are formed. This quasi particles are very short lived in metal nanoparticles, they are it is not like the electron hole pairs we create in the semiconductors. Semiconductors in semiconductors when we create electron hole pairs they are very long lived, we have seen that it can live up to picosecond time scale.

But, in metal nanoparticles electron hole pairs are created immediately after plasmon decay and that can be very short lived, within 10 femtosecond it can thermalize. That is why in this figure we have shown that within 10 femtosecond plasmon can decay to a electron non-thermal distribute distribution. And this non-thermal distribution is nothing, but having electron hole pair are created already and that can thermalize within 100 femtosecond time scale. Hot electron distribution features of Fermi Dirac distribution of electrons with well defined high temperature which is achieved mostly by electron-electron scattering.

The temperature scale, the temporal scale of these thermalization processes of few 100 of femtoseconds. Hot electrons further transfer energy to phonons of metal nanoparticles in a time scale of 1 to 5 picosecond. So, we have already mentioned that electron can transfer energy to phonon and that is very slow process 1 picosecond time scale. This heat transfer gradually increases lattice temperature depending on heat content. However, thermal diffusion can cool the hot lattice of the metal nanoparticles.

This cooling can eventually revert the photoexcited region back to the ambient value on the time scale of microsecond. So, metal nanoparticles can cool down in the time scale of microsecond, but these are the corresponding time cascaded processes which can be involved. And here we should also remember that although we are showing different steps in different time scale, but it may so happen that one step may overlap with the other step.

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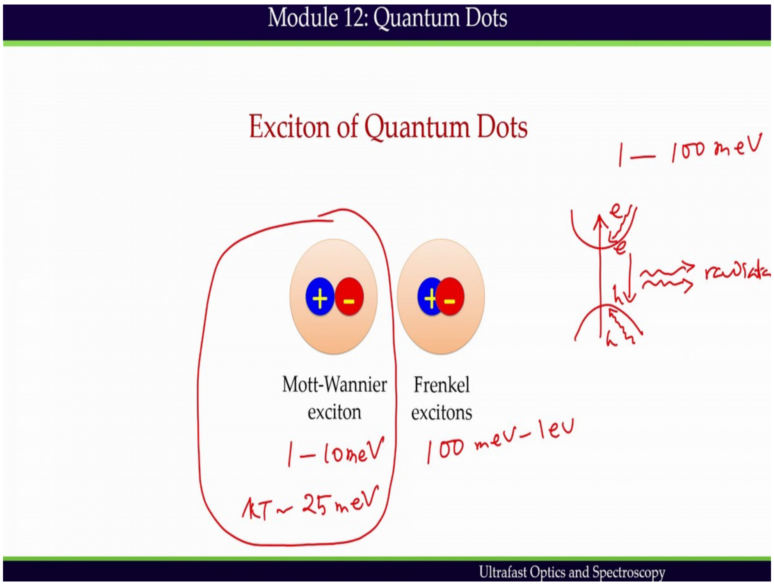

Module 12: Quantum Dots

Exciton of Quantum Dots

Mott-Wannier exciton

Frenkel excitons

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The last subject which we will cover in this module is ultrafast relaxation of quantum dots. Quantum dots are referred to semiconducting nanocrystals, they exhibit a profound size and shape dependent optical properties due to quantum confinement effect; upon absorption of a photon of light by a quantum dot an exciton is created. Exciton again it is an electron hole pair which is created and exciton in a quantum dot is an electron hole pair with associated columbic attraction.

So, the moment we create this electron and hole which is positive and negative charges, they must be associated with columbic attraction and this is called exciton. An illustration of strongly and weakly bound exciton counter dot is shown here, the exciton binding energy for most of the frequently encounter quantum dots is in the region in the range of a few to a few 10s of milli eV. So, it could be 1 to let say 100 milli eV, these excitons are called Mott-Wannier exciton.

The Mott-Wannier excitons are typical binding energy is found to be 1 to 10 milli eV, on the other hand Frenkel excitons typically exhibit much larger exciton binding energies. And the binding energy is almost 100 milli eV to 1 eV; Mott-Wannier exciton biding energies are usually small and comparable to the thermal energy kT which is nothing, but 25. This kT is 25 milli eV at room temperature and this is why it is observed that most excitons easily dissociate at room temperature.

Because, most of the inorganic quantum dots exhibit exciton binding energy on the order of in this region and that is why they can disassociate it. Exciton remains bound when the thermal energy is lower than the exciton binding energy. The study of exciton dynamics in quantum dots allows one to develop a deeper understanding of their fundamental properties. This can intern guide the design synthesis and interpretation of quantum dots for a wide variety of applications such as photocatalysis, photoelectrochemistry and photovoltaics.

Due to the relatively short lifetime of excitons, it is necessary to use ultrafast laser spectroscopy to probe or monitor directly their lifetime and associate relaxation or recombination processes. Dynamic processes in quantum dots are generally more complex than in the bulk due to the presence of many band gap states or surface states or trap states. Presence of band gap states can significantly alter exciton dynamics in quantum dots.

Some of the key and general dynamic processes are summarised here, in general photoexcitation of quantum dot with the light produces an excitation. In general photoexcitation of a quantum dot with light produces an exciton, an electron in the conduction band and a hole in the valence band; both of which are bound to each other by the force of Columbic attraction. Electron and hole can then undergo intraband relaxation with the electron relaxing to the bottom of the conduction band and hole to the top of the valence band.

So, something like which we have already shown here, we can excite during this excitation we create electron hole pairs excitons. And may so, happen that all electrons slowly will relax back to the bottom of the conduction band and hole can relax back to the top of the valence band. This relaxation occurs because of the electron phonon interactions and occurs on the time scale of several 10s to 100s of femtoseconds.

This relaxation occurs because of the electron-electron scattering and this occurs on the time scale of several 10s to 100s of femtoseconds. Once the electron reaches the bottom of the conduction band and hole reaches the top of the valence band, they may radiatively recombine. And when this recombination occurs then they can give radiation, emission; this is called photoluminescence. The electron hole pair can recombine non-radiatively with generally produces heat. Radiative recombination becomes prominent for near perfect semiconducting nanocrystals because, they exhibit a very low trap state density.

In this case the photoluminescence quantum yield becomes very high and the relevant time scale, relevant lifetime shows typically on the order of a few to a few 10s of nanosecond. However, for quantum dots with relatively high density of band gap states, trapping of electrons or holes into the states becomes important and occurs on time scale of a few picosecond to a few 10s of picoseconds. The time scale for trapping of charge carriers is usually faster than that of radiative recombination.

Following trapping charge carriers can either undergo traditional, following trapping charge carrier can either undergo additional trapping or recombine radioactively on or non-radioactively. The time scale for further trapping depends strongly on the nature of the trap states and can vary from picosecond to 100s of picoseconds. So, we have quickly gone over the different processes which can be involved in quantum dots, with this we have come to the end of this present module. In this module we have discussed different ultrafast processes which can be involved in solid, in semiconductor, in the metal nanoparticles, in quantum dots.

We will meet again for the next module.