Solar Photovoltaics:
Fundamental Technology and Applications
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Lecture-27
Photophysics of Perovskite Solar cells

Welcome everyone to our solar photovoltaics course 6th module, today we are having the 3rd lecture. We have already started with the perovskite solar cell, in the last 2 lectures we have discussed about the basic components of a perovskite solar cell. We discussed about what are the advantages of the perovskite solar cell and you also discussed how to fabricate a perovskite solar cell, what are the different components of perovskite solar cells.

In today's lecture we will discuss about the different kind of geometry or the orientation that is possible in a perovskite solar cell. And how this kind of geometry or the orientation can lead to the interesting photophysics in this perovskite solar cell.

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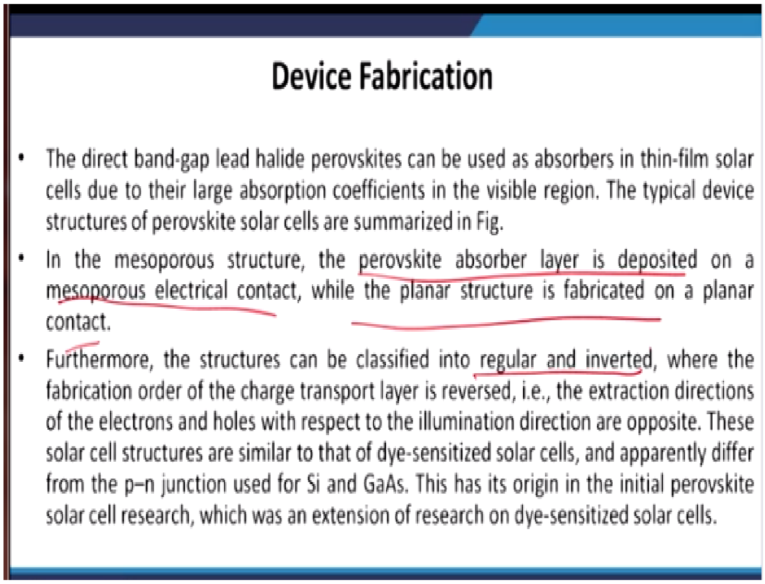

Device Fabrication

- The direct band-gap lead halide perovskites can be used as absorbers in thin-film solar cells due to their large absorption coefficients in the visible region. The typical device structures of perovskite solar cells are summarized in Fig.
- In the mesoporous structure, the perovskite absorber layer is deposited on a mesoporous electrical contact, while the planar structure is fabricated on a planar contact.
- Furthermore, the structures can be classified into regular and inverted, where the fabrication order of the charge transport layer is reversed, i.e., the extraction directions of the electrons and holes with respect to the illumination direction are opposite. These solar cell structures are similar to that of dye-sensitized solar cells, and apparently differ from the p–n junction used for Si and GaAs. This has its origin in the initial perovskite solar cell research, which was an extension of research on dye-sensitized solar cells.

So, device fabrication continued the direct band-gap lead halide perovskite can be used as absorbers in thin-film solar cells due to their large absorption coefficients in the visible regions. The typical device structure of perovskite solar cells we have already discussed and also have been shown in the subsequent figures. In the mesoporous structure the perovskite they absorber layer is deposited on a mesoporous electrical contact how can you get a mesoporous electrical contact.

So, what is the photo anode do you have, we have ITO coated glass substrate or FTO coated glass substrate as the photo anode. Now we mention that we can put a layer of titanium dioxide on top of that, now this TiO_2 layer we can make it to the mesoporous or a planar layer. So if we make a mesoporous TiO_2 structure by hitting the titanium dioxide in pH acetic acid solution after making the film at 450 degree Celsius we can make a mesoporous TiO_2 layer.

And if we use this mesoporous TiO_2 layer as a electrical contact then the resultant device will be a mesoporous photovoltaic devices. Similarly the planar structure is fabricated on a planar contact so if we do not want to make a corrugated porous structure. If you just have a planar TiO_2 layer then we will have a planar photovoltaic devices. Furthermore the structures can be classified into regular and inverted, where the fabrication order of the charge transport layer is reversed.

That is the extraction directions of the electrons and holes with respect to the illumination direction are opposite. So basically this thing we have already discussed we call them as a NIP geometry and PIN geometry. Some people are in some text book it is also called as regular and inverted structure. The solar cells structures are similar to that of dye-sensitized solar cells and apparently differ from the p-n junction used for silicon and gallium arsenide.

This has it is origin in the initial perovskite solar cell research which was an extension of a research on dye-sensitized solar cells yeah. So, these are very interesting story on that, so the when people were working on the dye-sensitized solar cell and it was very very popular, it routinely get efficiency of 8-10%. Now the first time the perovskite was used as sensitizer molecule when you say that a decent efficiency of 3.8% that time perovskite material was used as a sensitizer molecule in a DSSC geometry.

Later on it was found that not only as an observer but perovskite can also act like a hole transport material. Then people thought that ok why do not you use the same material as a sensitizer and as well as the hole transport material. So, that gives rise to the efficiency enhancement in the perovskite solar cell and then it was found that the efficiency is quite high even without doing any surface modification, with some surface modification we can get even a higher efficiency.

So, that is why all the people who are working on the sensitizer based solar cell they have also started working on perovskite solar cell.

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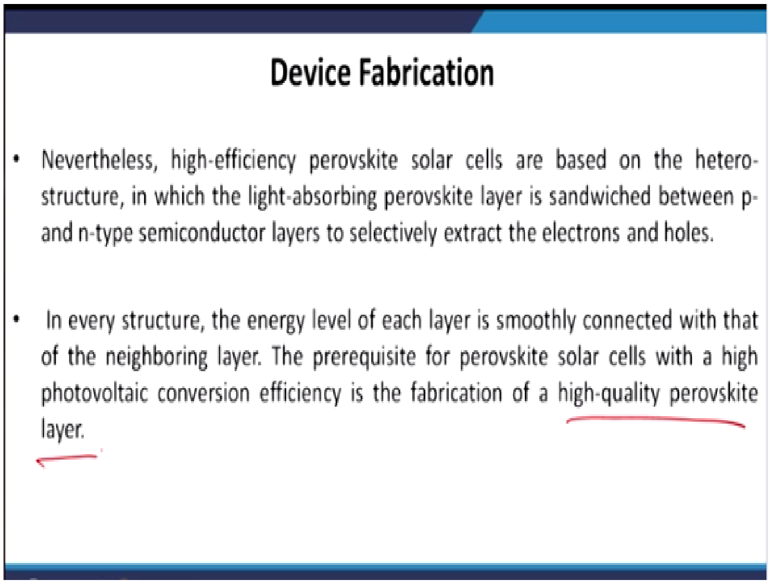

Device Fabrication

- Nevertheless, high-efficiency perovskite solar cells are based on the hetero-structure, in which the light-absorbing perovskite layer is sandwiched between p- and n-type semiconductor layers to selectively extract the electrons and holes.
- In every structure, the energy level of each layer is smoothly connected with that of the neighboring layer. The prerequisite for perovskite solar cells with a high photovoltaic conversion efficiency is the fabrication of a high-quality perovskite layer.

The high-efficiency perovskite solar cells are based on the hetero-structure, in which the light-absorbing perovskite layer is sandwiched between p and n-type semiconductor layers to selectively extract the electrons and holes. In every structure the energy level of each layer is smoothly connected with that of the neighboring layer. So, whenever we build up the successive layers we have to make sure about the energetics or we have to make sure about the position of their HOMO energy level and LUMO energy level.

So, that if proper electron transfer can happen in them, the perovskite the perquisite for perovskite solar cells with a high photovoltaic conversion efficiency is the fabrication of a high quality perovskite layer. This is somewhat similar to the organic solar cell you remember when we discuss about the organic solar cell we talked about the morphology. And we say that in an ideal organic solar cell we require a by continuous phase separate percolated network why by continuous phase separate it.

Because in organic solar cell we generate exciton bound electron hole pair and this excitons are generated in a donor material. We need to bring an acceptor material in the vicinity of the donor to separate the charge carriers to electrons and holes, so that electron goes in one way and hole goes in another way. Now we have to have a lot of the optimize nano interface throughout the geometry in a bulk hetero junction solar cell.

So that the charge carrier can hop from one site to the another site, now the physics of the perovskite solar cell is little bit the photophysics of the perovskite solar cell is somewhat different from the organic solar cell. Unlike excitons behavior here you have a free charge carrier behavior. Here the electrons and holes are not tightly bound by the electrostatic force they have free charge carriers, so we do not need to bring another material to separate the charge carrier.

But what is required, a very good quality a large uniform phenol free or crack free continuous thin films of the perovskite. So the phase separated by continuous percolated network in the organic solar cell is now replaced by a continuous in uniform phenol free large area perovskite films. If we have a continuous uniform and phenol free perovskite layer then the electrons and hole can easily percolate or transport using this layer.

But very often it is difficult to obtain a uniform layer with a very high quality and phenol free regions. So, that is comes in the domain of the morphology optimize and in the perovskite solar cell like before we will see that is morphology optimization relates to the photophysics of the devices. As you can understood that if the morphology optimization is not good if there are lot of phenols in the film, so this phenols acts like a trap states.

Now there can be 2 different kind of trap states or defect states in the perovskite layer, some of the trap states lies on top surface they are called cello trap states. And some of the trap states they are buried inside the bulk of the material they are called deep trap states. Now both these kind of trap states while cello trap on deep trap they act as a source of charge carrier recombination or they act like a place for charge carrier recombination.

So, in an ideal device we should not have lot of traps in the device, so we need to passivate those traps there are ways to passivate these traps or neutralize this traps by putting different material layers on top of the perovskite material. And by doing (()) (08:17) time florescence decay experiment, we can find out by doing an intensive dependant study how many trap or defectors are there in the perovskite layer.

Here they are located so that we can optimize this trap states to make a phenol free flims. So, as usual the morphologies always related to the photophysics of the device.

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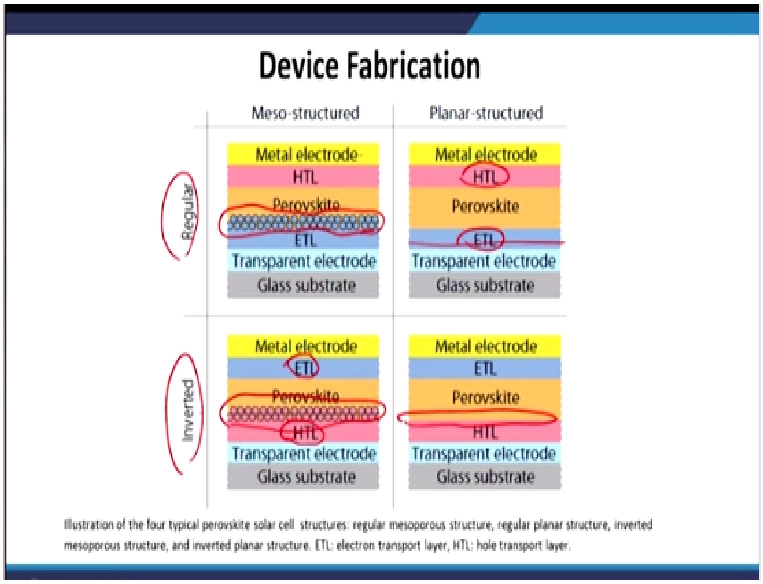

Device Fabrication

Illustration of the four typical perovskite solar cell structures: regular mesoporous structure, regular planar structure, inverted mesoporous structure, and inverted planar structure. ETL: electron transport layer, HTL: hole transport layer.

So let us take a look about the device fabrications, so if you remember we have talked about 2 different kind of geometry. One is the mesoporous structure another was the planar structure and you also talked about PIN or NIP geometry or regular or inverted geometry. Now it is possible to have a mesoporous or inverted mesoporous of the planar structure in either regular or in inverted, so based on that there are 4 different combinations of this perovskite solar cell is possible.

So for example you look at this picture here if you have a regular perovskite solar cell with a meso-structured orientations. So, I have a glass substrate transparent electrode ETL or electron transport layer, perovskite, hole transport layer and metal electrode, where are plain structure things with the regular plain structure glass substrate, transparent electrode, ETL, perovskite, HTL and metal electrode, so in both of this case it is a regular structure.

So, that is why I have the ETL electron transport layer with the ITO contact and hole transport layer at the metal contact. But in the first example is mesoporous so the TiO2 layer is mesoporous you can see from the figure we have drawn the circular things here. And here we did not draw any circular thing this is the plain things so one of them is a mesoporous layer another is a plain layer, on the other hand you look at the inverted structure here.

Here it will be a other way round glass substrate, transparent electrode, hole transport layer and electron transport layer in between the perovskite. So, what is the difference from these 2 figures here the electron transport layer was near the ITO but now hole transport layer is near the ITO. There the HTL was near the metal electrode now the ETL is near the metal electrode, so basically they have now inverted from the regular structure.

Now it can be also be mesoporous like this or it can be plain like this. So, there are 4 different configuration is possible regular meso-structured, regular planar structured, inverted meso-structured or inverted planar structured.

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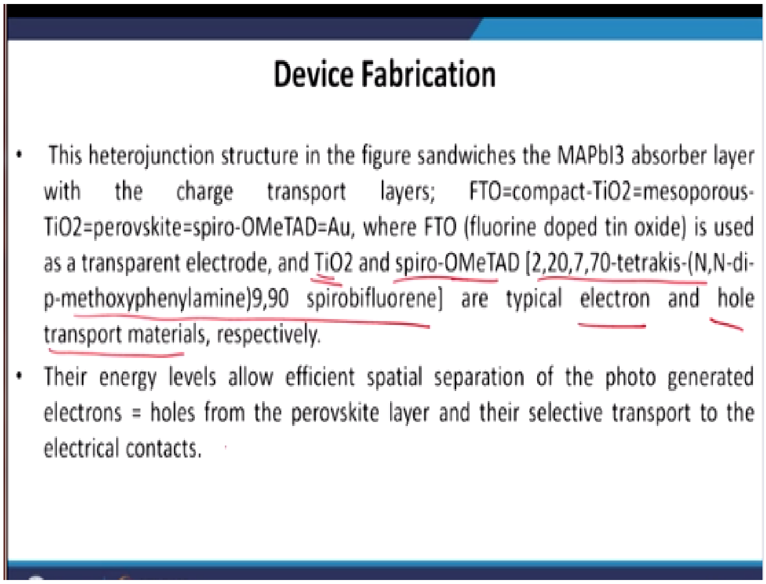

Device Fabrication

- This heterojunction structure in the figure sandwiches the MAPbl3 absorber layer with the charge transport layers; FTO=compact-TiO2=mesoporous-TiO2=perovskite=spiro-OMeTAD=Au, where FTO (fluorine doped tin oxide) is used as a transparent electrode, and TiO2 and spiro-OMeTAD [2,20,7,70-tetrakis-(N,N-di-p-methoxyphenylamine)9,90 spirobifluorene] are typical electron and hole transport materials, respectively.
- Their energy levels allow efficient spatial separation of the photo generated electrons = holes from the perovskite layer and their selective transport to the electrical contacts.

The heterojunction structure in the figure sandwiches the MAPbl3 absorber layer with the charge transport layers FTO compact TiO2 which can be mesoporous TiO2 perovskite and spiro-OMeTAD and finally the gold where FTO fluorine doped tin oxide is used as a transparent electrode and TiO2 and spiro-OMeTAD. They are typically as an electron and hole transport material. So TiO2 is a electron transparent material and spiro-OMeTAD which has this big name inside this bracket is a small molecule, this is a hole transport materials.

So it can be either an electron transport material and we are using a hole transport material along with our perovskite layer. Their energy levels allow efficient spatial separation of the photo generated electrons holes from the perovskite layer and their selective transport to the electrical contacts. So, they are not only help to separate the charge carriers but they also help to selectively transport the charge carriers to the corresponding photo anodes or photo cathodes or corresponding the photo electrodes.

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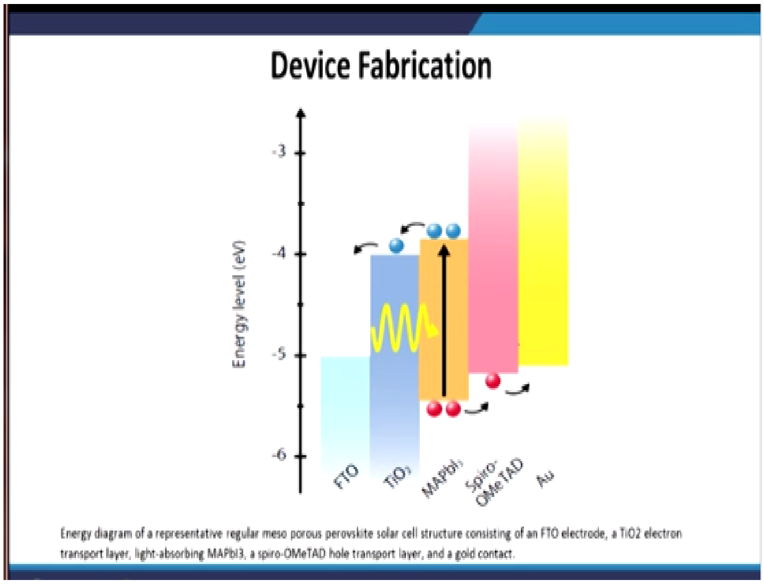

Device Fabrication

Energy diagram of a representative regular meso porous perovskite solar cell structure consisting of an FTO electrode, a TiO2 electron transport layer, light-absorbing MAPbI3, a spiro-OMeTAD hole transport layer, and a gold contact.

If you look the stacking of the devices we can also draw in terms of the energy band diagram. For example here you see that this is the FTO layer then on top of that there is a TiO2 layer then we have the perovskite layer MAPbI3 in top of that we have a spiro OMeTAD layer and finally the gold layer. Now find the light absorbs in the active layer so both the electrons and holes are created.

Electron is injected to the TiO2 because it is conduction band is lower than the LUMO energy level of the perovskite layer. And from there electron is injected to the FTO and finally it goes through the outside circuit. Whereas the hole which leaves here that goes to the spiro-OMeTAD from there it goes to the metal electrode, so this show the circuit completes.

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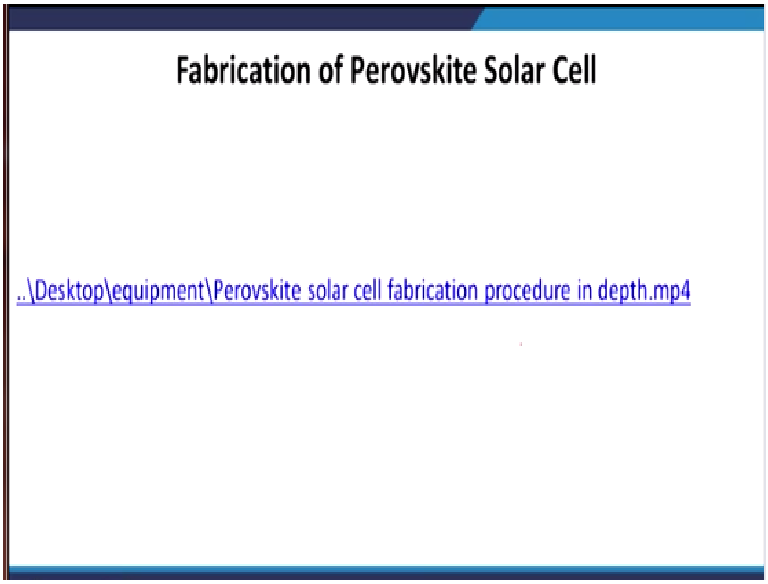

Fabrication of Perovskite Solar Cell

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Fabrication of perovskite solar cell we already discussed.

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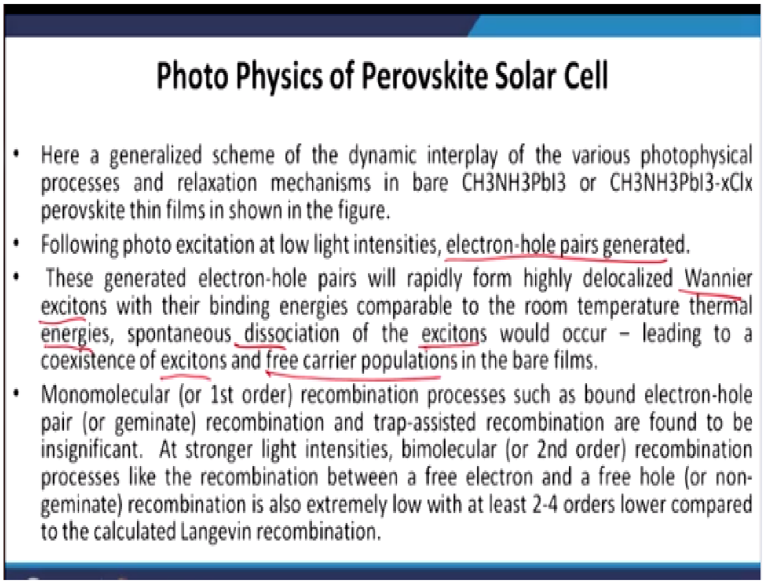

Photo Physics of Perovskite Solar Cell

- Here a generalized scheme of the dynamic interplay of the various photophysical processes and relaxation mechanisms in bare CH3NH3PbI3 or CH3NH3PbI3-xClx perovskite thin films in shown in the figure.
- Following photo excitation at low light intensities, electron-hole pairs generated.
- These generated electron-hole pairs will rapidly form highly delocalized Wannier excitons with their binding energies comparable to the room temperature thermal energies, spontaneous dissociation of the excitons would occur – leading to a coexistence of excitons and free carrier populations in the bare films.
- Monomolecular (or 1st order) recombination processes such as bound electron-hole pair (or geminate) recombination and trap-assisted recombination are found to be insignificant. At stronger light intensities, bimolecular (or 2nd order) recombination processes like the recombination between a free electron and a free hole (or non-geminate) recombination is also extremely low with at least 2-4 orders lower compared to the calculated Langevin recombination.

But what is most interesting thing is here when you discussed about the different orientation on the morphology is the photophysics of the perovskite solar cell. Later on we will see that there are lot of engineering aspect to optimize the morphology of the perovskite solar cell and each of them will lead to an interesting photophysics. So, what you mean by the photophysics of a perovskite photo solar cell.

Here a generalized scheme of the dynamic interplay of the various photophysical process and relaxation mechanisms in bare CH3NH3 lead iodide CH3NH3 lead iodide-x chlorine x perovskite thin films is shown in the figure. Following photo excitation at low intensity electron-hole pairs generated, so basically we generate electron hole pairs following photo excitation at low light intensity.

These generated electron-hole pairs will rapidly form highly delocalized wannier excitons with their binding energies comparable to the room temperature thermal energies. As a result spontaneous dissociation of the excitons should occur leading to a coexistence of excitons and free carrier populations in the bare films. So, the first process in any solar cell is the exciton generation or charge carrier generation.

So, similarly when light falls on the perovskite material electron-hole pairs are generated the first **is** is full-filled. Then the whatever electron-hole pair is generated there now delocalized and this kind of delocalized excitons are called 1 year excitons. If the excitons are very strongly bound and not delocalized there called frenkel excitons. And this 1 year excitons their binding energy is comparable to the room temperature thermal energies.

So if their binding energies comparable to the room temperature thermal energy what will happen so thermal energies enough energetic to break this binding energy of the excitons. So, it will spontaneously dissociate the excitons into the free charge carrier or the bound electron-hole pair will now dissociate to a free charge carrier. So in addition to the bound excitons we have now free carrier population in our perovskite film.

Monomolecular or first order recombination process such as bound electron-hole pair or geminate recombination and trap-assisted recombination are found to be insignificant. At stronger light intensity bimolecular or second order recombination process like the recombination between a free electron and a free hole or non-geminate recombination is also extremely low with at least 2 to 4 orders lower compared to the calculated by langevin recombination.

So, there are various kind of recombinations, recombination is the electron-hole pair annihilation. Now there can be various process of recombination happens, the recombination can be bound electron hole pair this is called the geminate recombination or the mono molecular recombination or the first order recombination. Now in perovskite this geminate recombination properties also low there can be trap assisted recombinations.

So, there the existence of this geminate recombination and the trap assisted recombination but if you have a very uniform continuous film or a very uniform continuous phenol free film. Then you do not have lot of trap assisted recombination. So that is why both the band to band recombination or the geminate recombination as well as trap assisted **re** recombination is insignificant for this perovskite material.

Now when we increase the intensity of the light if we increase the intensity of the light then the second order recombination that becomes very very dominant. Second order recombination is a free electron hole recombination or non-geminate recombinations. Now the probability of non-geminate recombination is also low at least 2 to 4 order lower compared to the calculated langevin recombination model in the perovskite material.

Now because of this low recombination we will have an excellent charge carriers transport to the electrodes and that is leads to the high short circuit current in this kind of device.

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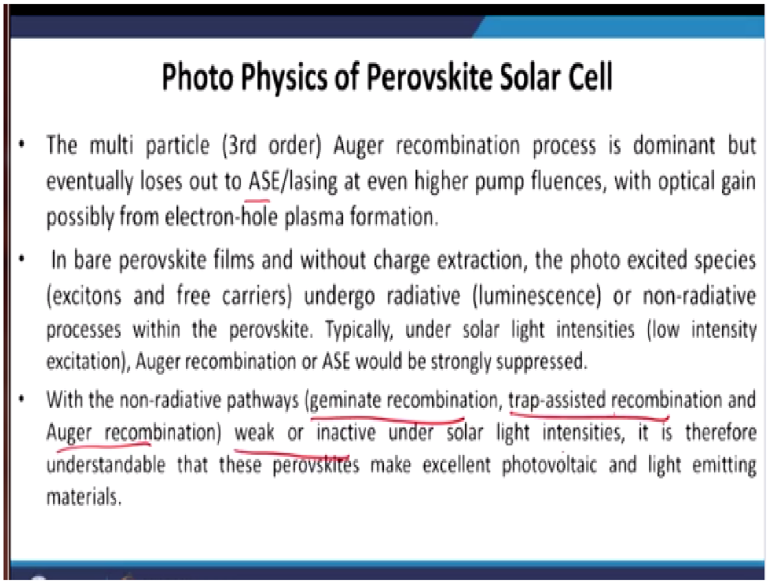

Photo Physics of Perovskite Solar Cell

- The multi particle (3rd order) Auger recombination process is dominant but eventually loses out to ASE/lasing at even higher pump fluences, with optical gain possibly from electron-hole plasma formation.
- In bare perovskite films and without charge extraction, the photo excited species (excitons and free carriers) undergo radiative (luminescence) or non-radiative processes within the perovskite. Typically, under solar light intensities (low intensity excitation), Auger recombination or ASE would be strongly suppressed.
- With the non-radiative pathways (geminate recombination, trap-assisted recombination and Auger recombination) weak or inactive under solar light intensities, it is therefore understandable that these perovskites make excellent photovoltaic and light emitting materials.

Now what left is the multi particle third order auger recombination process, so the multi particle recombination is now dominant. But eventually loses out to amplified stimulate emission or ASE or lasing at even higher pump fluence with optical gain possibly from the electron-hole plasma formation. So, there are 3 different kind of recombination can happen in any opto-electronics devices ok.

So, the first is the first order recombination sometimes called the geminate recombination this is due to the band to band recombination or bound electron hole pair recombination, there can be trap assisted recombination. If you go to the high intensity of the light then the second order recombination becomes dominant. Second order recombination is the free electron-hole recombination this is also called a non-geminate recombination.

And there are third order recombination or multi particle auger recombination now in the case of the perovskite both the first order and second order recombination is insignificant. But due to the presence of the exciton and the free charge carrier simultaneously the multi-particle auger recombinations become prevalent in this kind of system. But finally what happens this multi-particle recombinations they lead to the ASE or lazing phenomena at high pump fluence intensity.

In bare perovskite films and without charge extraction, the photo excited species excitons and free carriers undergo radiative luminescence or non-radiative process within the perovskite. Typically under solar light intensities at low intensity Auger recombination or ASE would be strongly suppressed. So if I use the low intensity excitation then the Auger recombination is not very strong effect.

But if I use the high intensity radiation then the Auger recombination become dominant. Experimentally by doing a femtosecond time resolved spectroscopy one can find out the different components of this recombinations, from the decay dynamics of the excited charge carriers. One can find out whether it is a first order recombination, second order recombination or Auger recombination. With the non-radiative pathways geminate recombination, trap-assisted recombination and Auger recombination weak or inactive under solar light intensities.

It is therefore understandable that these perovskites make excellent photovoltaic and light emitting materials. Now what are the source of the non-radiating pathways first of all they geminate recombination or first order band to band recombination it can be trap-assisted recombination and it can be Auger recombination or multi-particle recombination. Both these geminate recombination or trap-assisted recombination or Auger recombination they are all non-radiative pathways.

So, that means they do not emit the light, so that is why they are weak or inactive under solar light intensities and therefore if the non-radiating emissions is there or if the non-radiating pathways are weak. So then the radiating pathways has to be very very strong that is why this kind of direct band gap materials are also suitable for fabrications not only for photovoltaic devices but also for the light emitting diodes.

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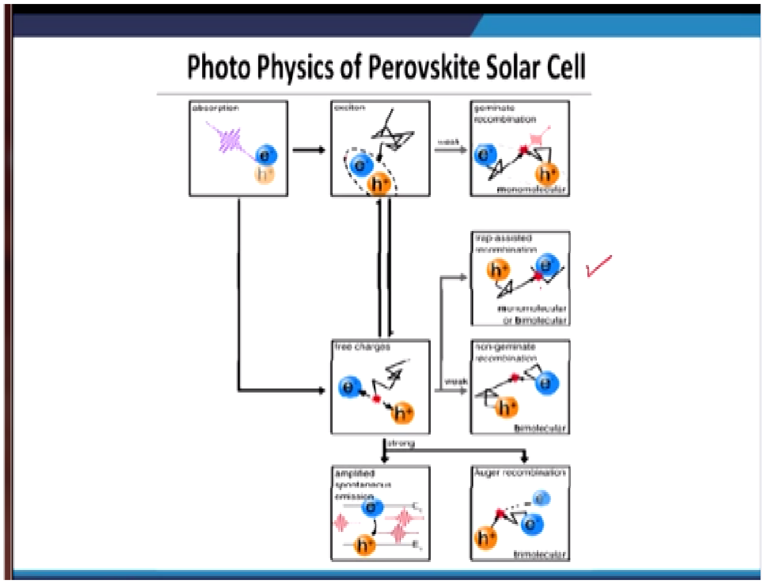

Photo Physics of Perovskite Solar Cell

Let us take a look of this band diagram or a schematic figure of the hole photo physics of a perovskite solar cell. So what happens so when light absorbs we create an electron hole pair now this bound electron hole pair we call this as an exciton. Now if the exciton they are weakly bound if this an 1 year excitons is an weakly bound then we see a geminate recombination. So, the electron can go and couple with the hole is a mono-molecular recombination.

Similarly these excitons these probabilities very very weak this exciton can disintegrate to the free charge carriers. So, electron and holes is now free and they can wander around using this acting material geometry. Now 2 case can be possible between this free electron hole pair if there is an weak intensity of the light then there will be an non geminate recombination or second order recombination, free charge carrier, free electron hole recombination or there can be trap-assisted recombination.

These electrons and hole when they travel along the films if they are defect states or trap-states is there like cello trap states or deep trap states then electrons and hole can call upon on those trap states and these trap states are a potential energy minima so they can get recombine there. So the free charge carriers can undergo 2 different kinds of recombination, non-geminate recombination which is happens under the weak intensity of the light or it can be trap-assisted recombination.

Now if the morphology of the film is very good then we can discard this trap-assisted recombination exciton under the weak intensity they got the geminate recombination that means band to band recombination. Now if you have a strong intensity of the light then what will happen in amplifieds spontaneous emission or multi-particle Auger recombination, in a multi-particle Auger recombination it is a tri-molecular recombination.

So the electron-hole can combine with another electron or here there can be further amplified emission is here. So to summarize this whole process once again absorbance of the light they generate a bound electron hole pair but this electrons and hole pairs are nor bound loosely. So there are 2 things happen either this bound electron hole pair can disintegrates to a free charge carrier free electrons hole or they can under the weak intensity of the light band to band geminate recombinations and this is a monomolecular recombinations.

Now when we have a free charge carrier decay then the free charge carrier they can undergo 2 different kinds of recombination. Under the weak intensity of the light they can be an non-geminate recombination which is the free electron hole recombination, it is a second order recombination or there can be trap-assisted recombination. If there are lot of trap states or lot of defect states in the material then that can be tarp-assisted recombination.

If the morphology of the perovskite film is very good then we can suppress this root of the recombination. Now the third possibility is that if the light intensity is very very strong then there can be spontaneous amplified emission or can be Auger recombination or multi-particle recombinations. Now all these 3 different mechanisms can happen from the perovskite material now depending upon the morphology we can suppress one of this root.

Now if there electron-hole pair is recombine then what will happen if the electron-hole pair is recombine radiatively then we will get the light. If it is recombine non-radiatively then we will get the heat. So, all the source of non-radiative pathways as we discussed in the last slide if you go back to the last slide all the source of the non-relative pathways like geminate recombination the first order recombination, trap-assisted recombination, free charge carrier recombiantions and the Auger recombination.

The third order recombination under the strong intensity of the light, these are all non-relative pathways. First order geminate recombination, trap-assisted recombination and Auger recombinaitons, these are all non-relative source of the light. That means this kind of recombination does not give the light they are weak. So which is strong is the bimolecular recombination or non-geminate recombination in the perovskite.

And if there is a bimolecular or non-geminate recombination is there, then we will get a radius amount of the light out of the circuit. So that is why the photophysics tells us like you know these materials is good for fabricating light emitting diodes also. Except the fact like in although like in a what you learn that this photo physics is excellent in the perovskite material but there are some problems also with the perovskite materials.

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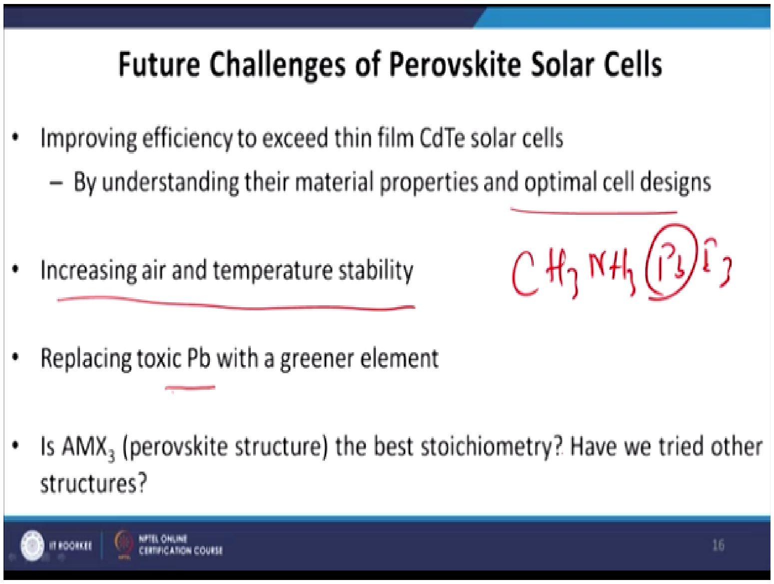

Future Challenges of Perovskite Solar Cells

- Improving efficiency to exceed thin film CdTe solar cells
  - By understanding their material properties and optimal cell designs
- Increasing air and temperature stability
- Replacing toxic Pb with a greener element
- Is AMX_3 (perovskite structure) the best stoichiometry? Have we tried other structures?

$CH_3NH_3\boxed{Pb}I_3$

For example some of the challenge of the perovskite solar cells are improving efficiency to exceed thin film cadmium telluride solar cell. Now thin film cadmium telluride solar cell as already achieved like 16 to 18% or even like you know 20% efficiency in an ideal condition and which are very very stable. But perovskite solar cell although has reach that number but the stability is very very poor.

So by understanding that material properties and optimal cell designs one can improve the efficiency. But the most important challenge is the increasing air and temperature stability, we will discuss in details later on about this air and temperature stability and how we can prevent this instability. Because there are lot of research interest is now ongoing and to make a stable air passable ambient conditions stable perovskite solar cell.

Replacing the toxic lead with a greener element, now we said that when you have this perovskite materials CH3NH3 lead iodide we have lead in this material but lead is toxic. Although we use lead in the battery but still there is a possibility of the environmental leaching, so we do not want lead to have in the systems. so that is why lot of research interest is ongoing to replace the lead with non-lead material for example bismuth or silicon, so these are called lead free peroxide.

One of the problem with the lead free peroxide is their efficiency is not very very high although there stability is very high in compression to the less based device. So the current challenge is to improve the lead free peroxide solar cells efficiency. Finally is the AMX3 perovskite structure the best stoichiometry, have we tried other structures. There are lot of interest is now going instead of the single perovskite structure to make a double perovskite structure or a layered perovskite structures which is called the quasi perovskite 3D, 2D perovskite structures.

Or a 2D structure or like you know try a completely different kind of stoichiometry or the geometry in the perovskite structure and see why whether that is also good enough for the charge carrier transport. So these are all challenge in the fields which needs to be optimized for a better understanding of the photophysics of the device and as you have seen that in many cases the morphology and photo physics they are correlated.

If we have a very good morphology then we have a control over our photo physical properties even if we have a good materials. And some of the current challenge like the stability and the toxicity of the lead is addressed by developing the methods to counteracts the photo instability, thermal instability as well as the moisture instability. In our next lecture we will discuss about the stability issue of the perovskite solar cell which is one of the major issue which prevents the commercialization of this devices.

And then we will see that once we address the photo instability issue then we can also address the thermal instability and moisture instability and it is possible to make a air passable completely stable perovskite solar cell with a descent 8 to 10% efficiency even higher than that. So the possibility of making roll to roll perovskite solar module is not very far array and if it is possible then the dream of making the solar cells at a extremely low cost in a roll to roll flexible substrate can be achieved.

Now whatever we have discuss today regarding the photophysics of the perovskite as I said that lot of research papers and review papers excellent research papers and review papers if you search online you will get it.

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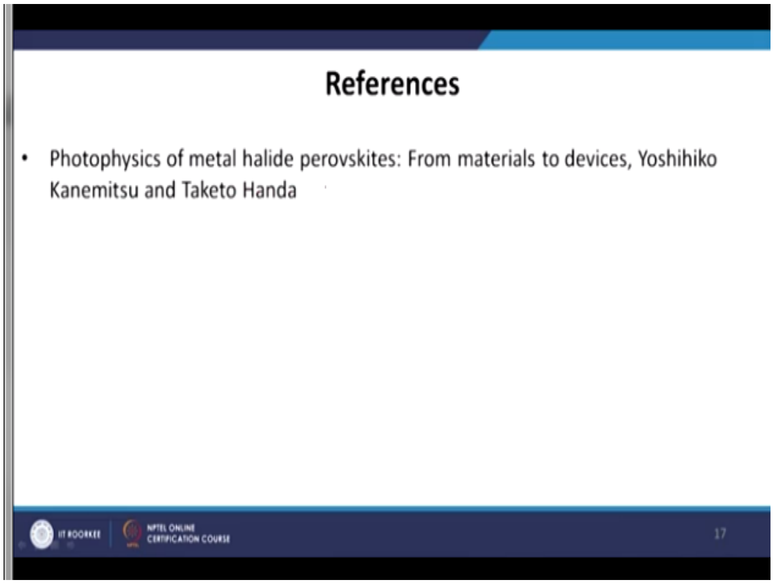

References

- Photophysics of metal halide perovskites: From materials to devices, Yoshihiko Kanemitsu and Taketo Handa

For example you can also this photophysics of metal halide perovskite from materials to devices by Yoshihiko Kanemitsu and Taketo Handa. This is a very good reference to look into and understand the basic concept of the photophysics of the perovskite. But there are lot of this very excellent review papers on the photophysics of the perovskite material which one can also look for better understanding, thank you for your attention.