First rare earth ions doped Si based down converter layers integration in an industrial Si solar cell

J. Cardin[1], L. Dumont[1], C. Labbé[1], P.-M. Anglade[1] , M. Vallet[2], M. Carrada[2], S.C. Wu[3], I.-S. Yu[3], A. L. Richard[4], W. M. Jadwisienczak[4], X. Portier[1], P. Marie[1], F. Gourbilleau[1]

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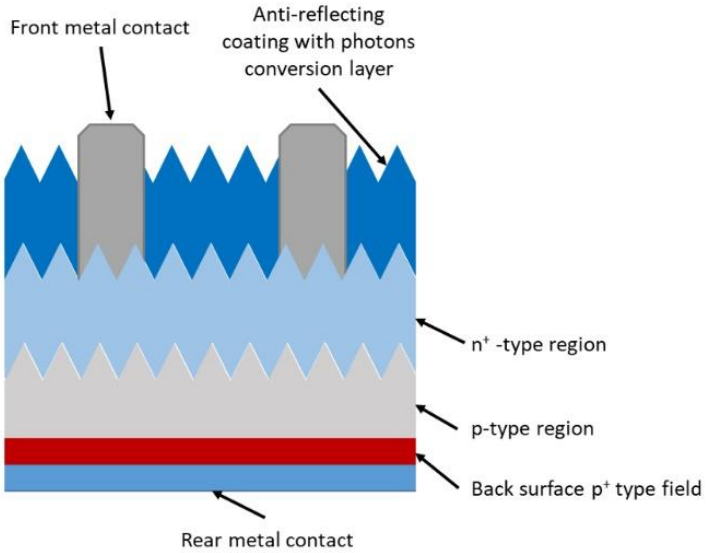
[3] *Department of Materials Science and Engineering, National Dong Hwa University, Hualien, Taiwan*

[4] *Ohio University, Athens, OH 45701, USA*

7th International Conference "Nanotechnologies and Nanomaterials" NANO-2019, 27 - 30 August 2019 in Lviv Polytechnic National University Lviv, Ukraine

Outline

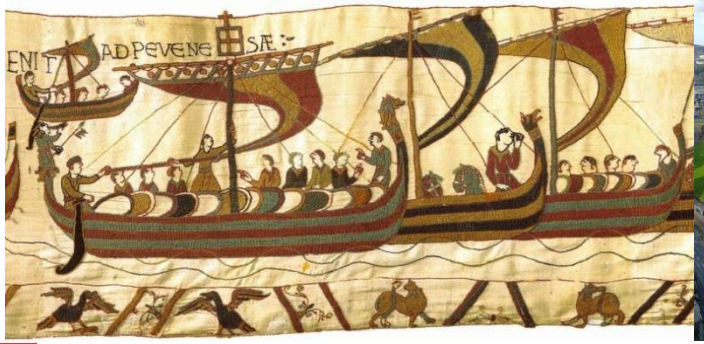
- Introduction
- Growth of thin films by RF magnetron sputtering
- Frequency layer optimization
- Modeling of Donor (Tb^{3+})-Acceptor(Yb^{3+}) ions system
- Integration in an industrial process
- Conclusion



Introduction

CIMAP Lab

In Caen, Normandie (2h with car in the west from Paris)



University of Caen Normandie, Ensicaen, CNRS, CEA

7 teams with various activities : irradiation with heavy ions, optical materials for laser and photonics, material for energy, defects studies in SC + biosourced materials…+ **CIRIL platform leading the researches** at the GANIL facility

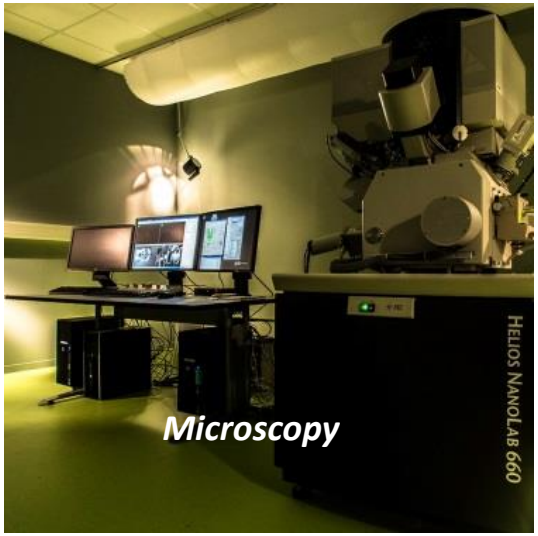
AMA MADIR PM2E **NIMPH** CIRIL-GANIL OML LARIA SIMUL

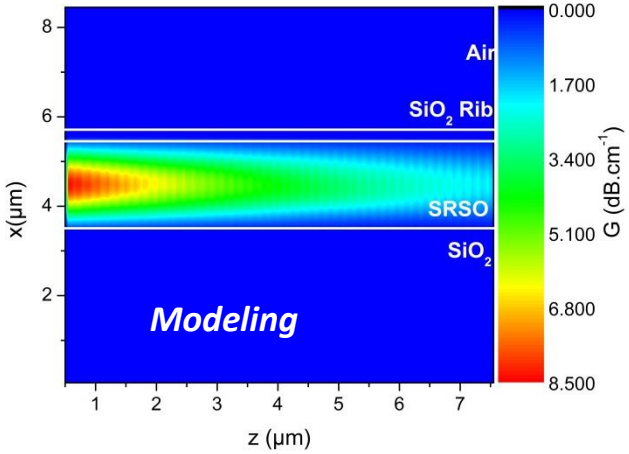
NIMPH Team


Atomic Layer Deposition

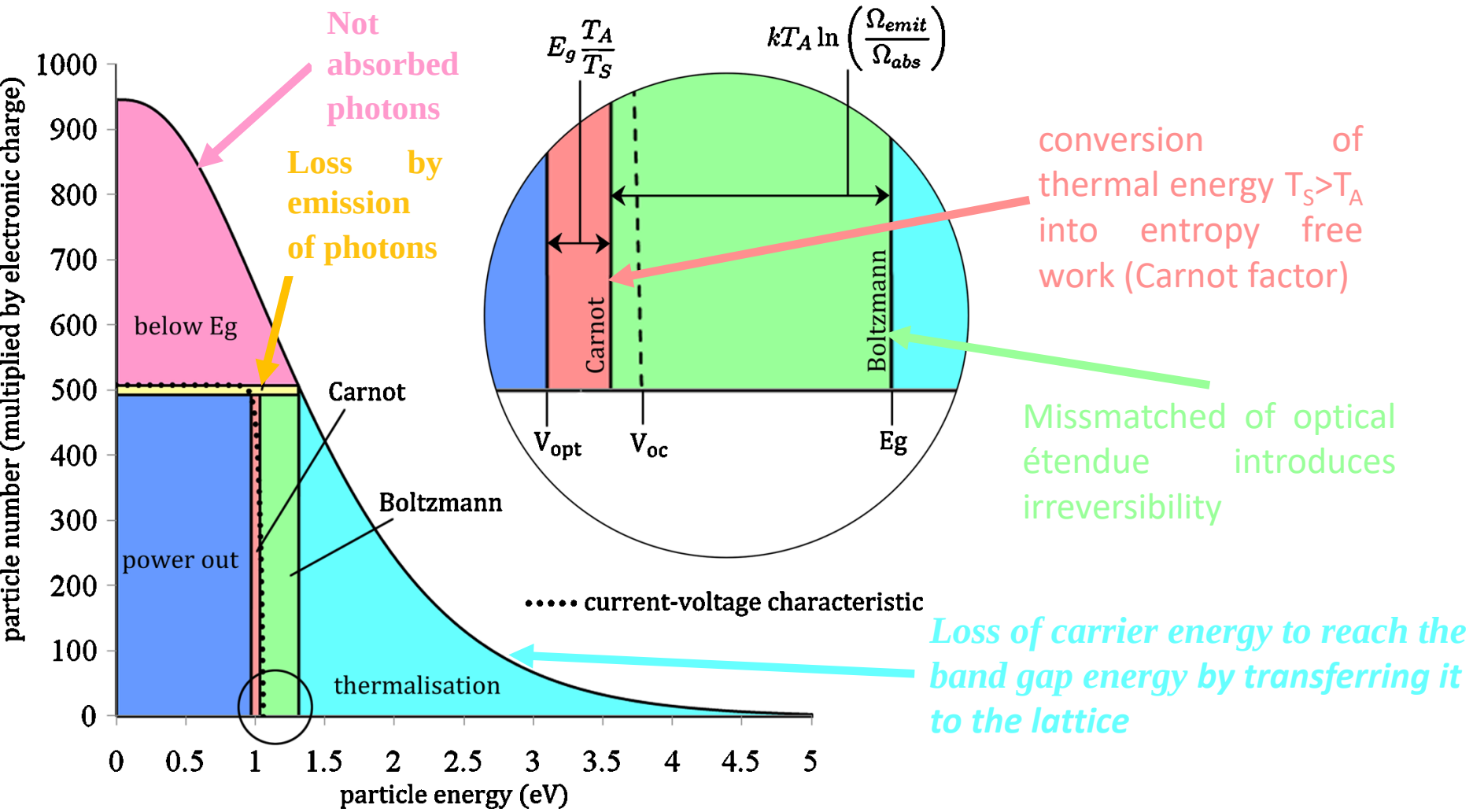

Magnetron sputtering


Optical/electrical carcterizations


Microscopy
HELIOS NANOLAB 660


Air
SiO2 Rib
SRSO
SiO2
Modeling
x(μm)
z (μm)
G (dB.cm-1)
0.000
1.700
3.400
5.100
6.800
8.500

Losses in solar cells



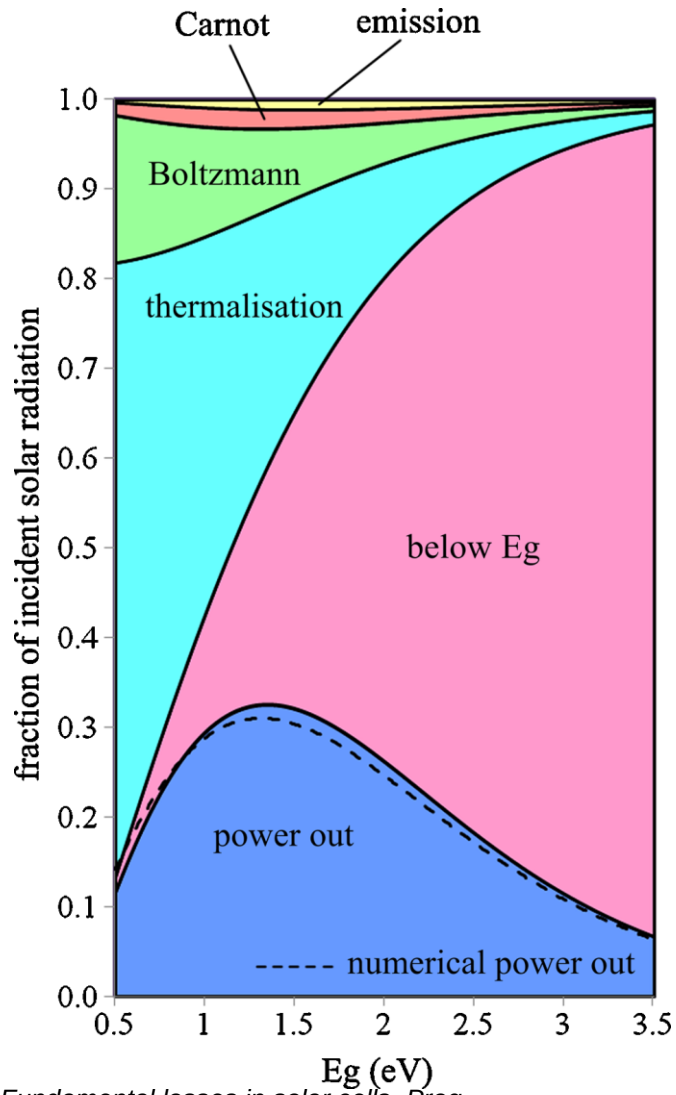
conversion of thermal energy $T_S > T_A$ into entropy free work (Carnot factor)

Missmatched of optical étendue introduces irreversibility

Loss of carrier energy to reach the band gap energy by transferring it to the lattice

*Fundamental losses in solar cells, Prog. Photovolt: Res. Appl., LC Hirst, N J Ekins-Daukes, **19**:286–293, 2011*

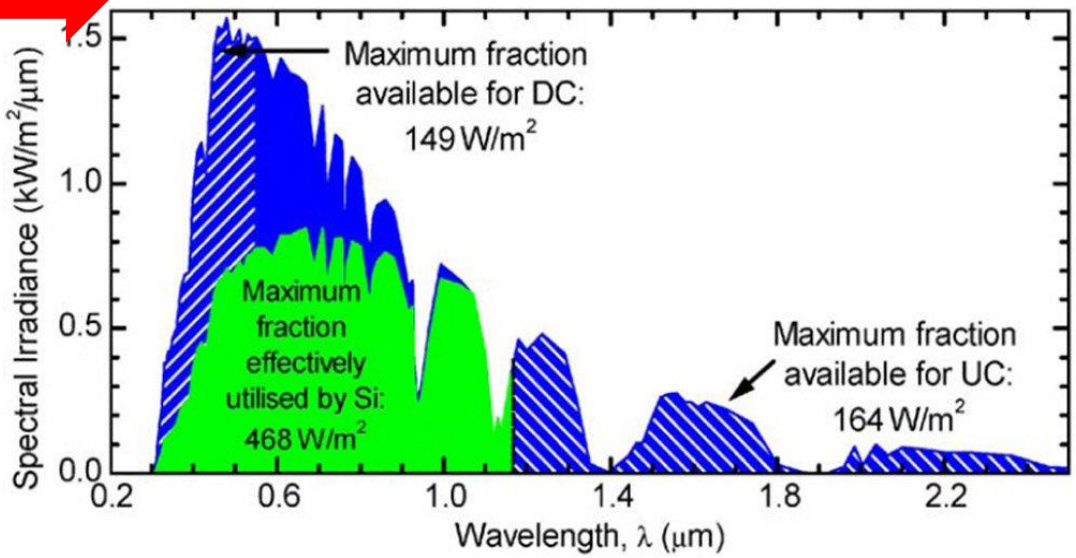
Losses in Solar Cells : limited efficiency



*Fundamental losses in solar cells, Prog. Photovolt: Res. Appl., LC Hirst, N J Ekins-Daukes, **19**:286–293, 2011*

Different kind of losses:

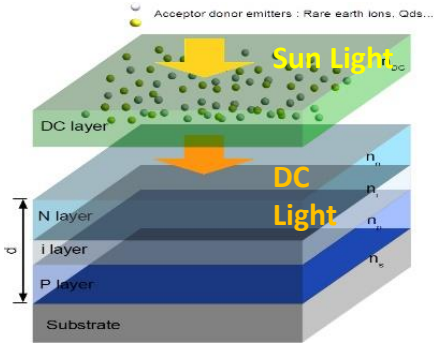
- Emission : recombination of carrier->PL
- Carnot limit : unavoidable heat loss to the surroundings
- Boltzmann : irreversible entropy generation
- Thermalization : carrier with energy largely above SC Eg
- Below SC Eg: photon with energy bellow SC Eg

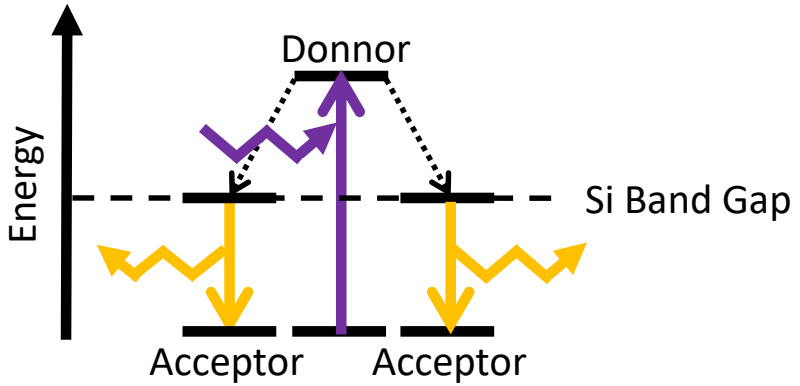


Yao Zhu et al, 7 January 2013, SPIE Newsroom. DOI: 10.1117/2.1201212.004585

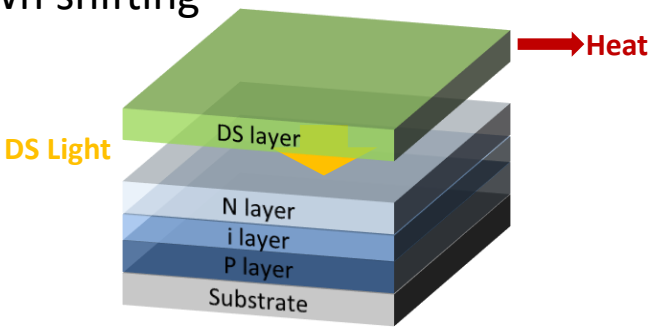
Solar irradiance conversion with solar cell

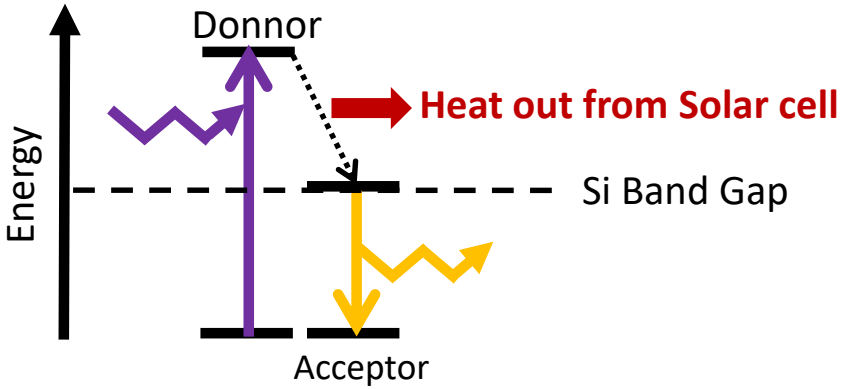
Down conversion



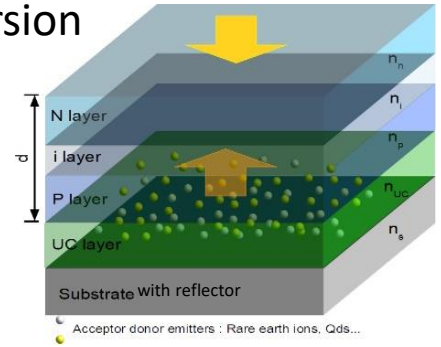


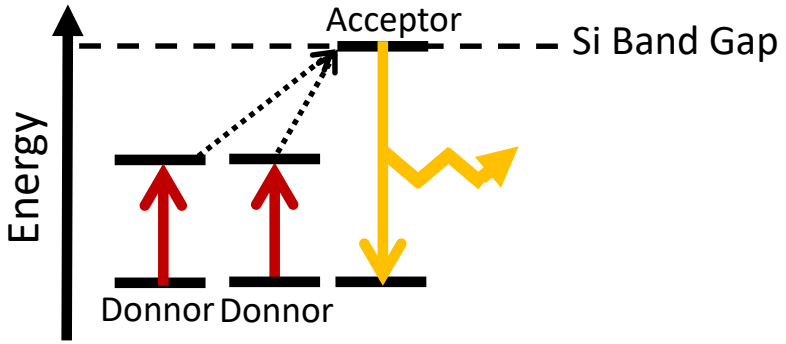
Down shifting



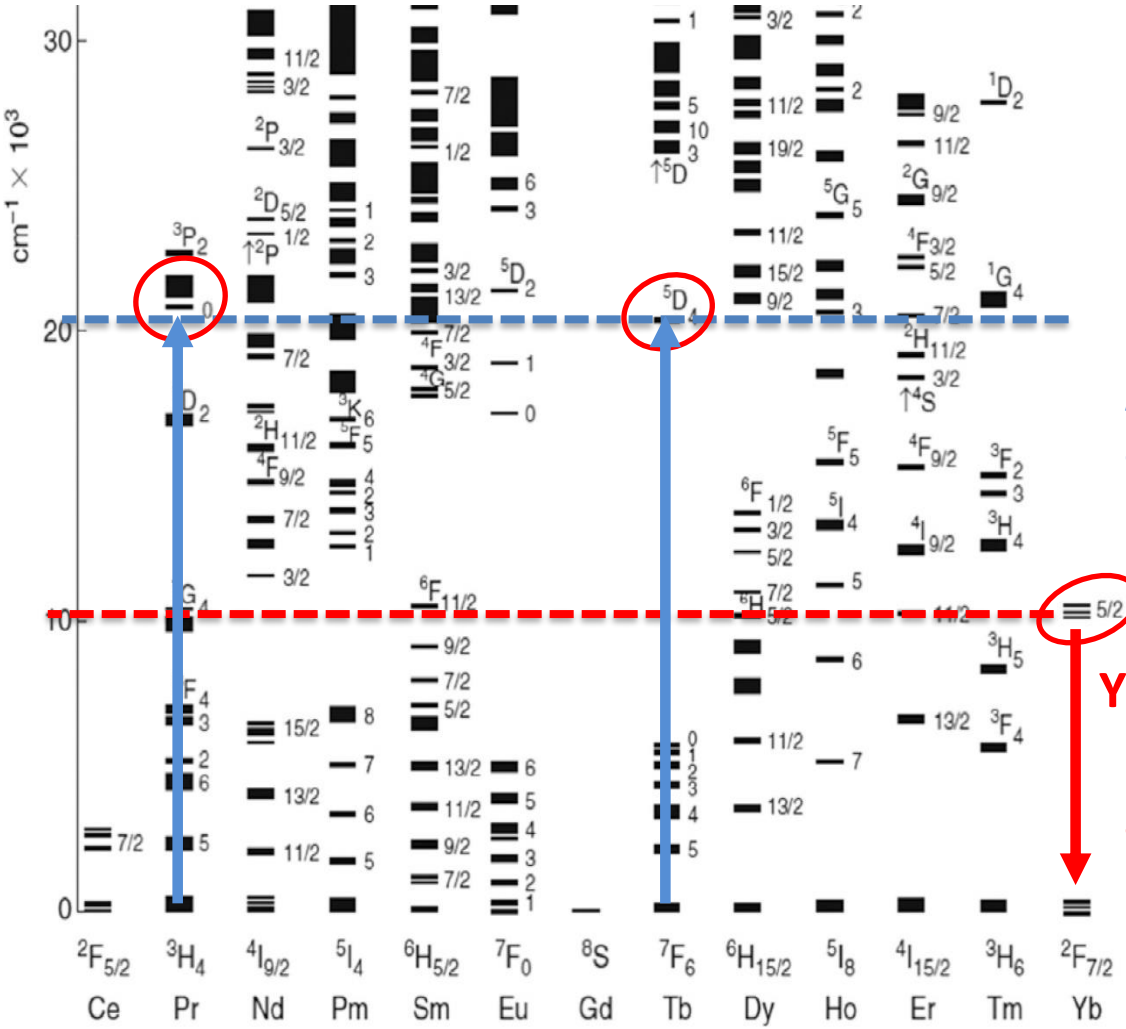


Up conversion





Rare earth ions for donnor-acceptor system



Pr^{3+} , Tb^{3+}

Absorption at about 500nm (2,48eV)
-> twice the SC BG

Yb^{3+}

Emission at about 1000nm (1,24eV)
-> once the SC BG

G. H. Dieke and H. M. Crosswhite, Applied Optics Vol. 2, Issue 7, pp. 675-686 (1963)

Development of frequency conversion layer

ADVANCED OPTICAL MATERIALS

Full Paper

Highly Efficient Infrared Quantum Cutting in Tb^{3+}–Yb^{3+} Codoped Silicon Oxynitride for Solar Cell Applications

Yong-Tao An, Christophe Labbé, Julien Cardin, Magali Morales, Fabrice Gourbilleau

First published: 10 September 2013

DOI: 10.1002/adom.201300186

SiN_xO_y:Tb^{3+}:Yb^{3+} efficient QC for frequency conversion layer

2013

2017 SiN_x:Tb^{3+}:Yb^{3+} efficient DS layer

Solar Energy Materials and Solar Cells

Volume 169, September 2017, Pages 132-144

ELSEVIER

Down-shifting Si-based layer for Si solar applications

L. Dumont [a], P. Benzo [a, 1], J. Cardin [a], I.-S. Yu [b], C. Labbé [a], P. Marie [a], C. Dufour [a], G. Zatryb [c], A. Podhorodecki [c], F. Gourbilleau [a]

2016

Solar Energy Materials and Solar Cells

Volume 145, Part 2, February 2016, Pages 84-92

ELSEVIER

SiN_x:Tb^{3+}–Yb^{3+}, an efficient down-conversion layer compatible with a silicon solar cell process

Lucile Dumont [a], Julien Cardin [a], Patrizio Benzo [a, 1], Marzia Carrada [b], Christophe Labbé [a], Andrea L. Richard [c], David C. Ingram [c], Wojciech M. Jadwisienczak [d], Fabrice Gourbilleau [a]

2016 SiN_x:Tb^{3+}:Yb^{3+} efficient DC layer

2017

Received: 13 April 2018 | Revised: 6 July 2018 | Accepted: 30 July 2018

DOI: 10.1002/pip.3071

RESEARCH ARTICLE

WILEY PROGRESS IN PHOTOVOLTAICS

First down converter multilayers integration in an industrial Si solar cell process

Lucile Dumont[1] | Julien Cardin[1] | Christophe Labbé[1] | Cédric Frilay[1] | Pierre-Matthieu Anglade[1] | Ing-Song Yu[2] | Maxime Vallet[3] | Patrick Benzo[3] | Marzia Carrada[3] | Didier Stiévenard[4] | Hocine Merabet[5] | Fabrice Gourbilleau[1]

SiN_x:Tb^{3+}:Yb^{3+} efficient Multilayer DC layer topped on Si solar cell.

2019

Journal of Rare Earths

Available online 7 November 2018

In Press, Accepted Manuscript

ELSEVIER

Monolithic crystalline silicon solar cells with SiN_x layers doped with Tb^{3+} and Yb^{3+} rare-earth ions ☆

Ing-Song Yu [a], Shao-Chun Wu [a], Lucile Dumont [b], Julien Cardin [b], Christophe Labbé [b], Fabrice Gourbilleau [b]

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https://doi.org/10.1016/j.jre.2018.07.014 Get rights and content

SiN_x:Tb^{3+}:Yb^{3+} based efficient DC layer topped on SI solar cell with an relative increase by 1.34% in the conversion efficiency

PhD YT An

PhD L Dumont

Growth of thin films by RF magnetron sputtering

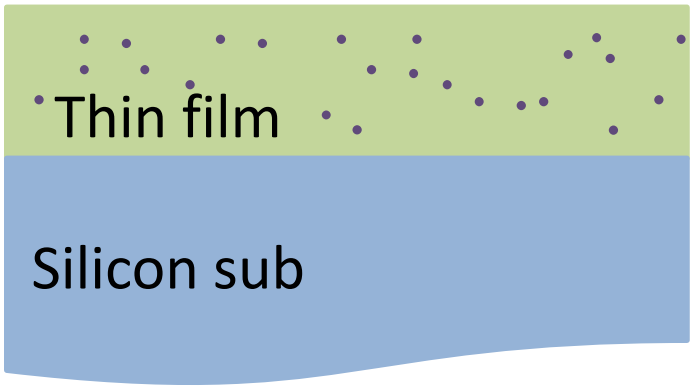
Important parameters

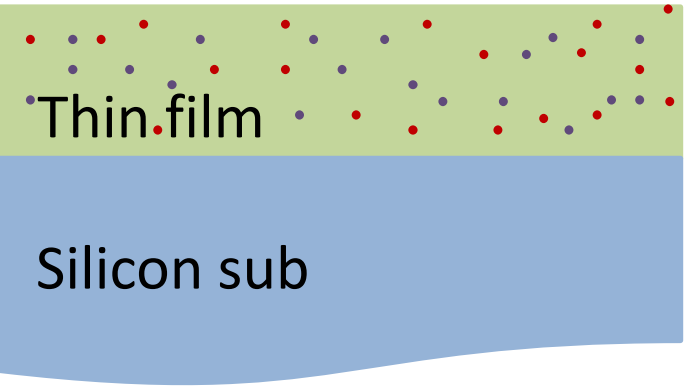
- **Chemical :**
  - rare earth (RE) ratio
  - activation of rare earth(RE^{3+})
  - high concentration required
- **Physical :**
  - energy matching between host matrix density of states and RE states
  - RE low excitation cross-section (10^{-21} cm^2)
  - Cross relaxation between same kind of RE
  - Excitation of Donor ions directly or indirectly (matrix sensitization)->Matrix (SiO_x, SiN_x, SiO_xN_y) properties
  - Cooperative energy transfer from donor (D) to acceptor (A) ion->D-A distance dependent
  - Relaxation mechanisms trough energy transfer between the same kind of ions->distance dependent
- **Optical :**
  - transparent in 550-1200nm range and efficient in UV (DC) or in IR (UC)
- **Technological Process :**
  - Compatible with Si solar cell process
  - stable in time (20-25years), non toxic

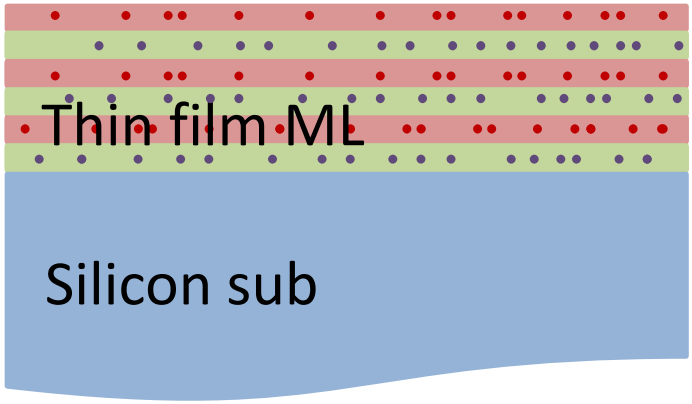
Si based down shifting and down conversion layers in homogenous and multi layer configurations

- SiO_y
- SiN_xO_y
- SiN_x

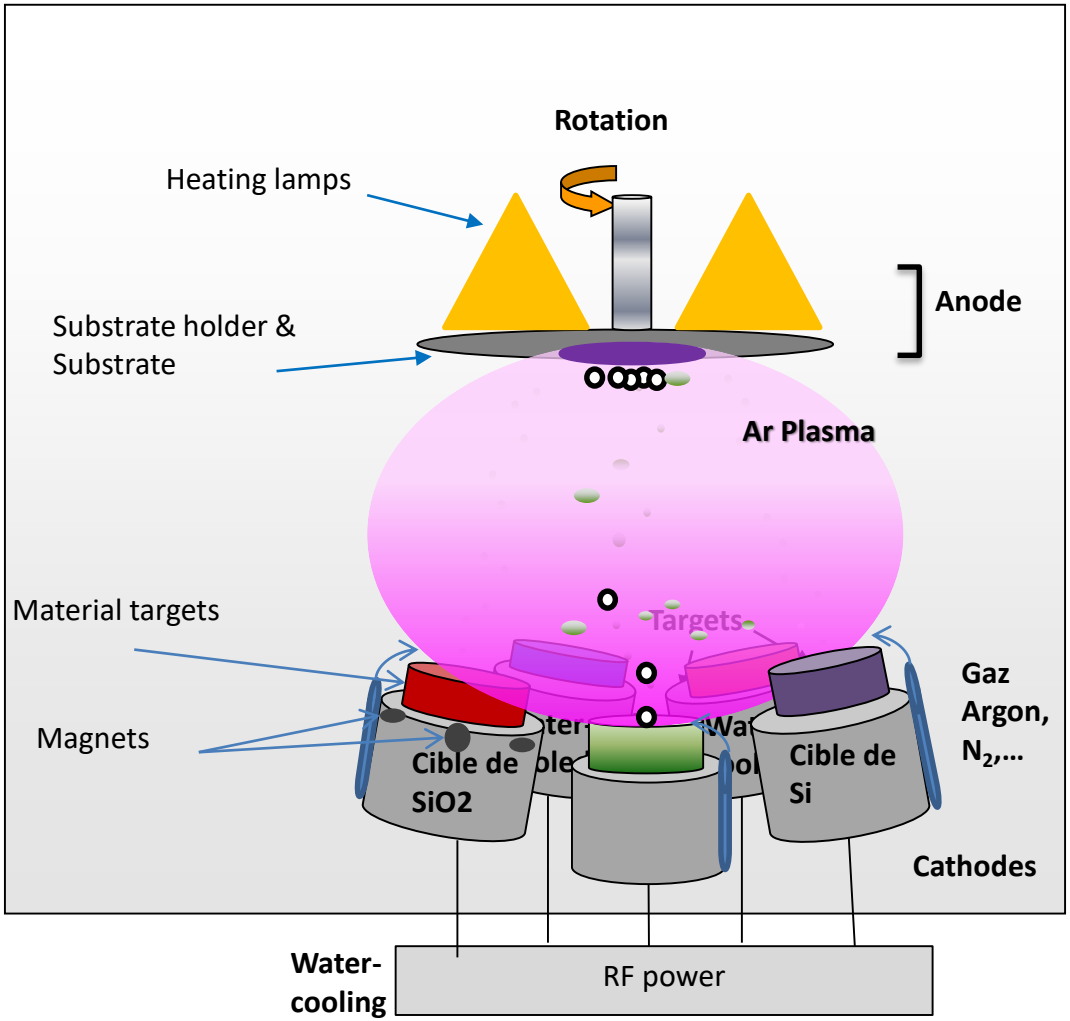
: $Tb^{3+}Yb^{3+}$





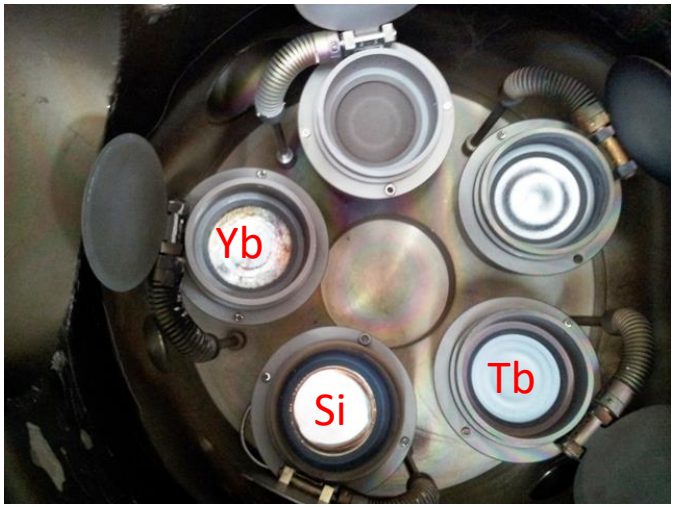


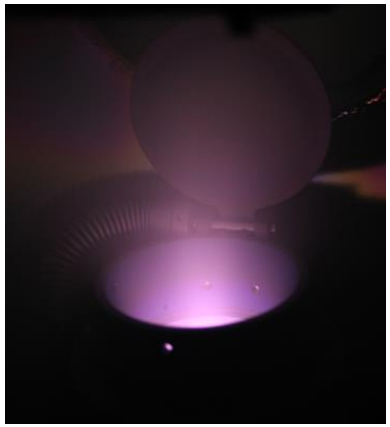
Growth of thin films by RF magnetron sputtering



Grown of thin film by magnetron sputtering

- Depositions parameters:
- Deposition Temperature , $\mathbf{T_{dp}}$ (°C)
- Plasma Pressure, $\mathbf{P_{tot}}$ (sccm)
- Gaz : Argon and nitrogen
    - Nitrogen rate, $\mathbf{rN_2}$
- RF Power density on target, $\mathbf{P_{target}}$ (W/cm^2)
- Depositing time, $\mathbf{t_{dp}}$ (s)



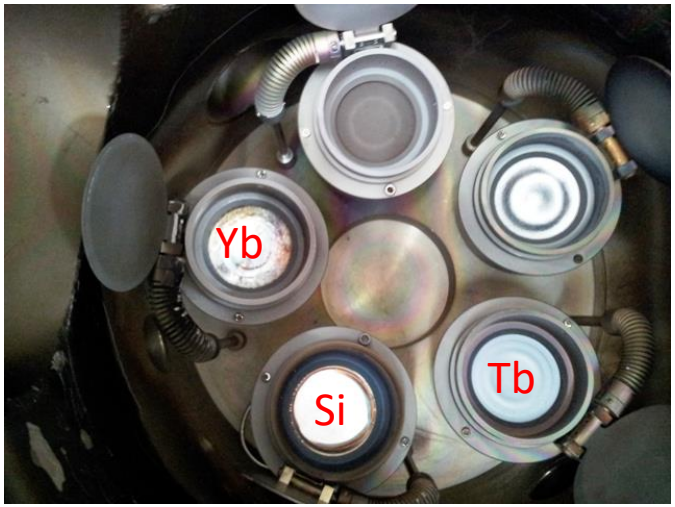


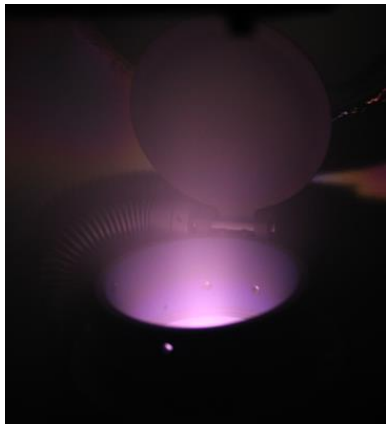
Confocales targets conf.

Plasma

Grown of thin film by magnetron sputtering

- Depositions parameters:
- Deposition Temperature , $\mathbf{T_{dp}}$ (°C)
- Plasma Pressure, $\mathbf{P_{tot}}$ (sccm)
- Gaz : Argon and nitrogen
    - Nitrogen rate, $\mathbf{rN_2}$
- RF Power density on target, $\mathbf{P_{target}}$ (W/cm²)
- Depositing time, $\mathbf{t_{dp}}$ (s)
- **Annealing with rapid or slow ramp of T under specific gas (N_2, Forming…)**

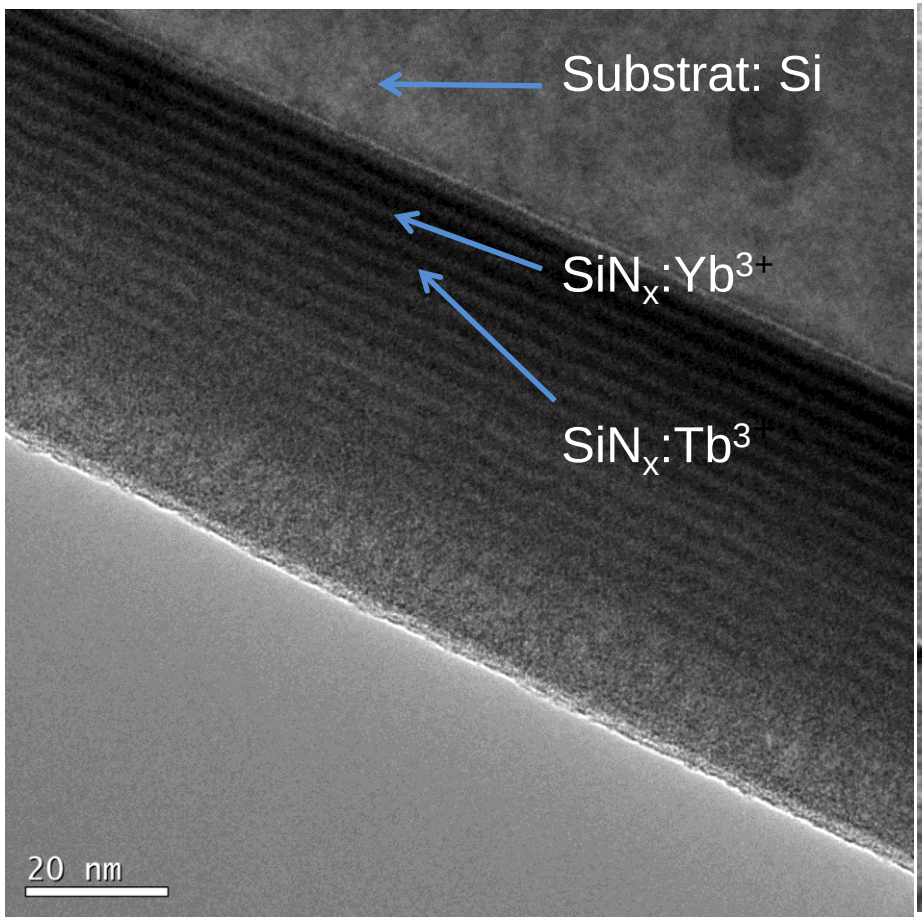




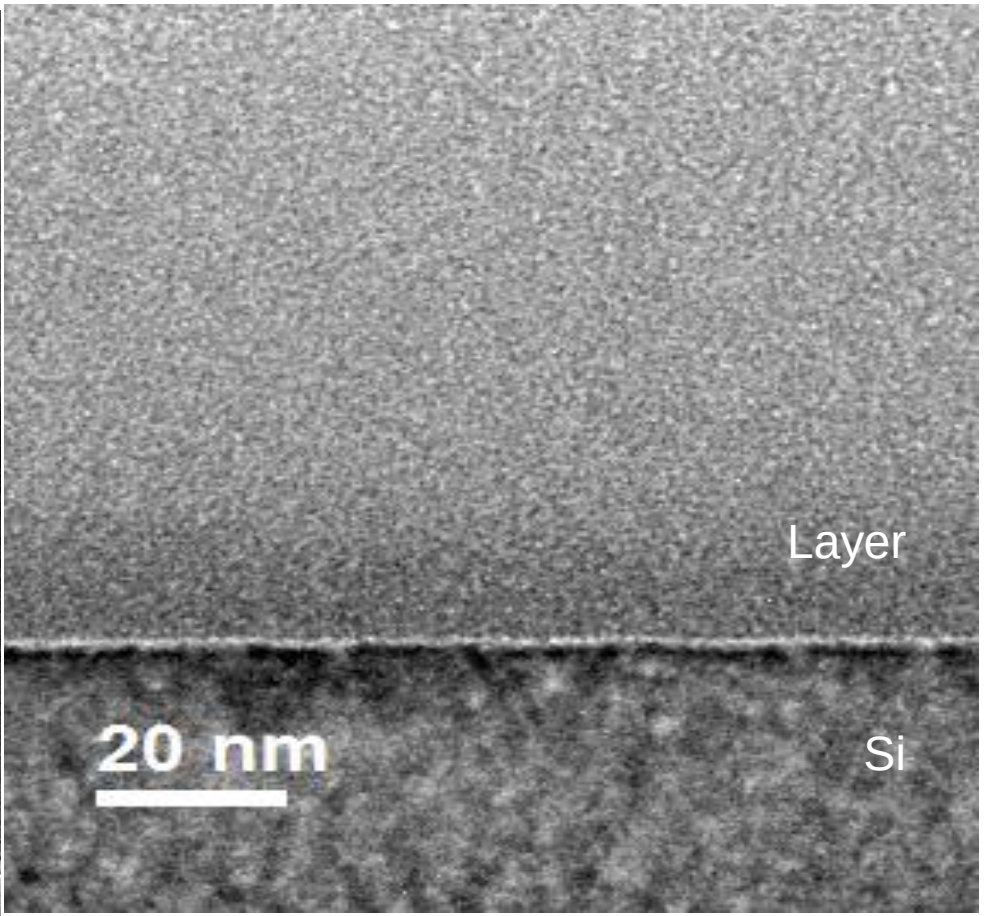
Confocales targets conf.

Plasma

Homogenous thin film and multilayer systems

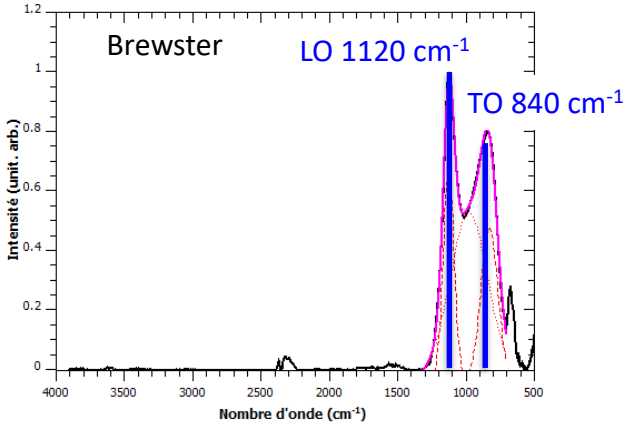


Cross-section view of the optimized multilayer system.

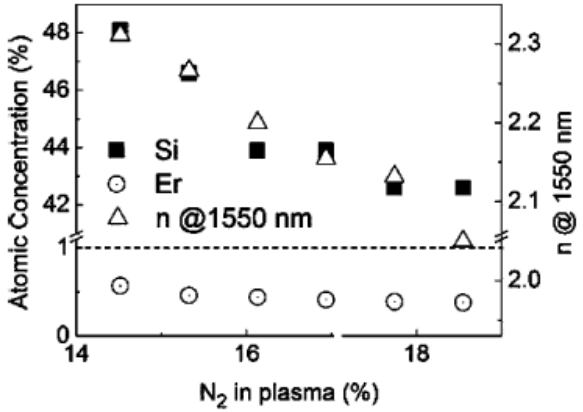


Cross-section view of the optimized homogenous layer system

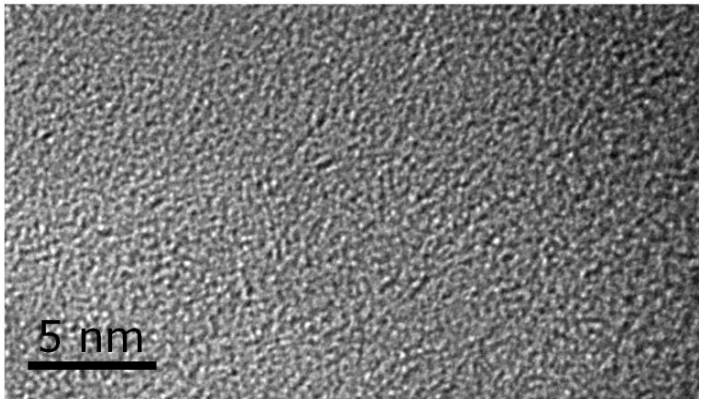
Structural and physical properties of Si based layer grown by magnetron sputtering



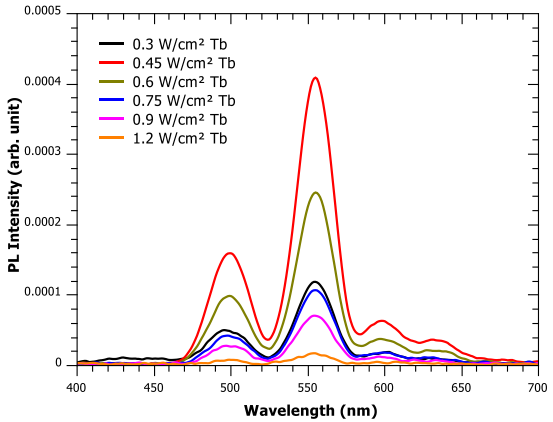
Control of the stoichiometry by tuning the sputtering parameters: SiN_x [FTIR]



Control of refractive index by tuning the sputtering parameters : SiN_x [ELLIPSOMETRY]



Amorphous and homogenous films: SiN_x [HRTEM]



Si based doped with rare earth ions luminescent layer: SiNx-Tb^{3+}[PHOTOLUMINESCENCE]

Frequency layer optimization

Down conversion layer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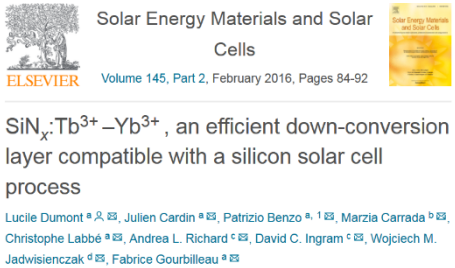
Full Paper

Highly Efficient Infrared Quantum Cutting in Tb^{3+}–Yb^{3+} Codoped Silicon Oxynitride for Solar Cell Applications

Yong-Tao An, Christophe Labbé, Julien Cardin, Magali Morales, Fabrice Gourbilleau

First published: 10 September 2013 Full publication history

DOI: 10.1002/adom.201300186 View/save citation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Solar Energy Materials and Solar Cells

Volume 145, Part 2, February 2016, Pages 84-92

SiN_x:Tb^{3+}–Yb^{3+}, an efficient down-conversion layer compatible with a silicon solar cell process

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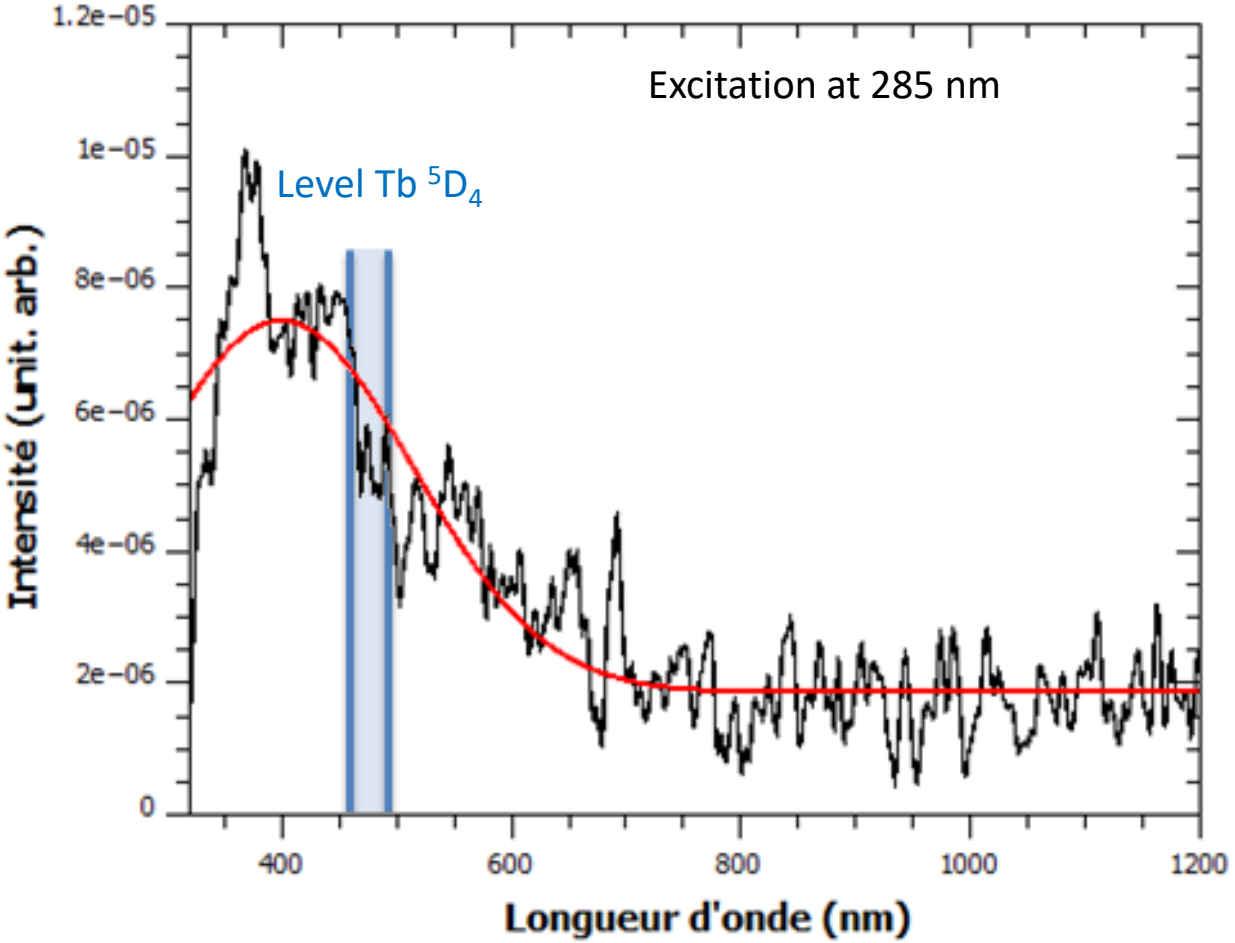
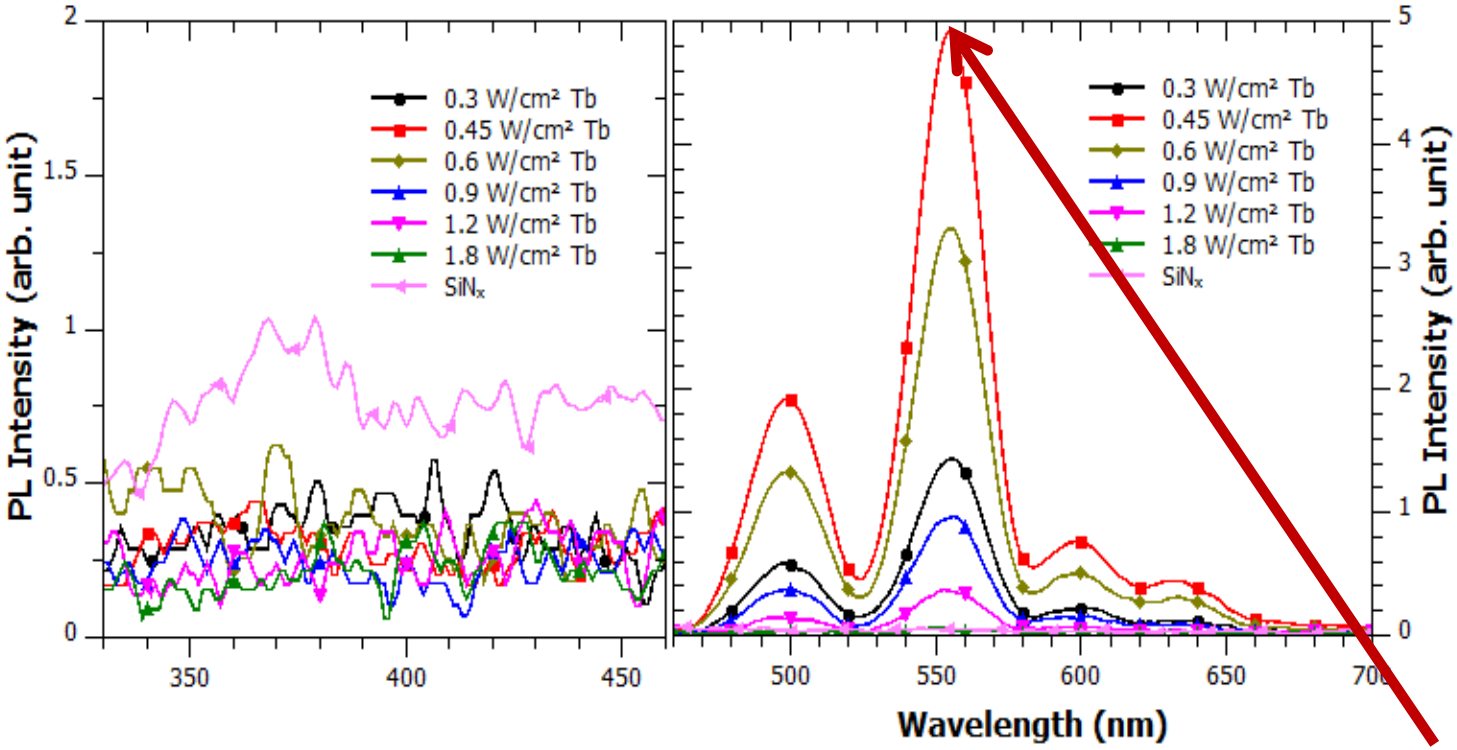
SiN_x matrix photoluminescence



Matrix Emission around 400nm
→ fit with Tb^{3+} absorption by level $^5D_4$ ➡ energy transfer may occurs

Energy transfer to Tb^{3+}



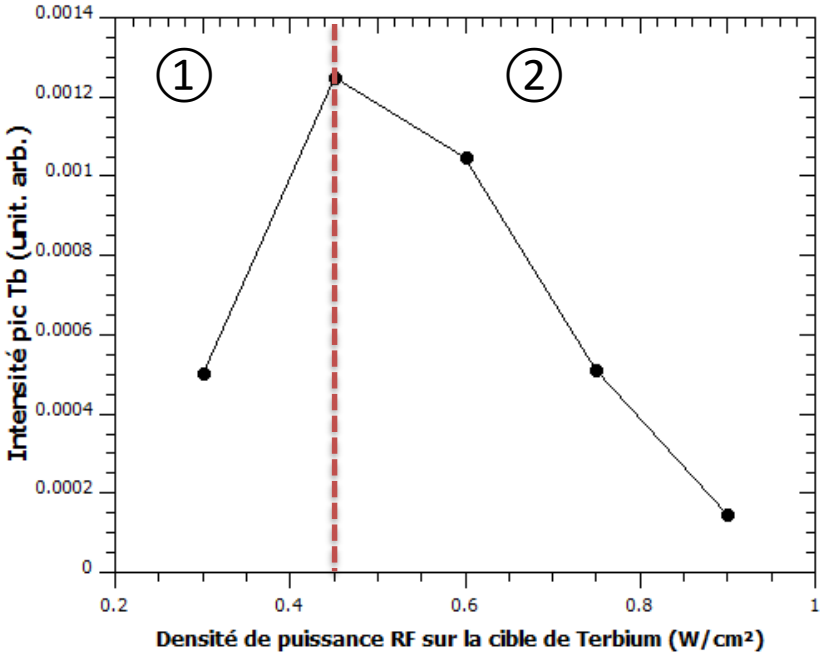
- Extinction of SiN_x matrix luminescence
- Increase of Tb^{3+} luminescence with Tb content

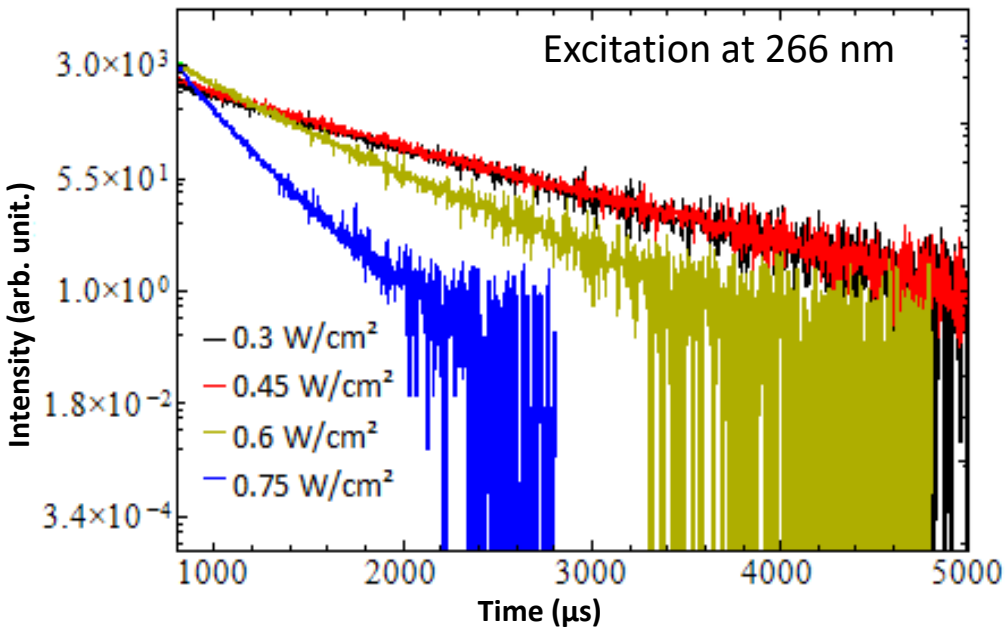
Optimum at 0,45 W/cm²

→ Indirect excitation of Tb^{3+} ions

→ Energy transfer (ET) from matrix to Tb^{3+} ions

Tb^{3+}concentration effect on luminescence

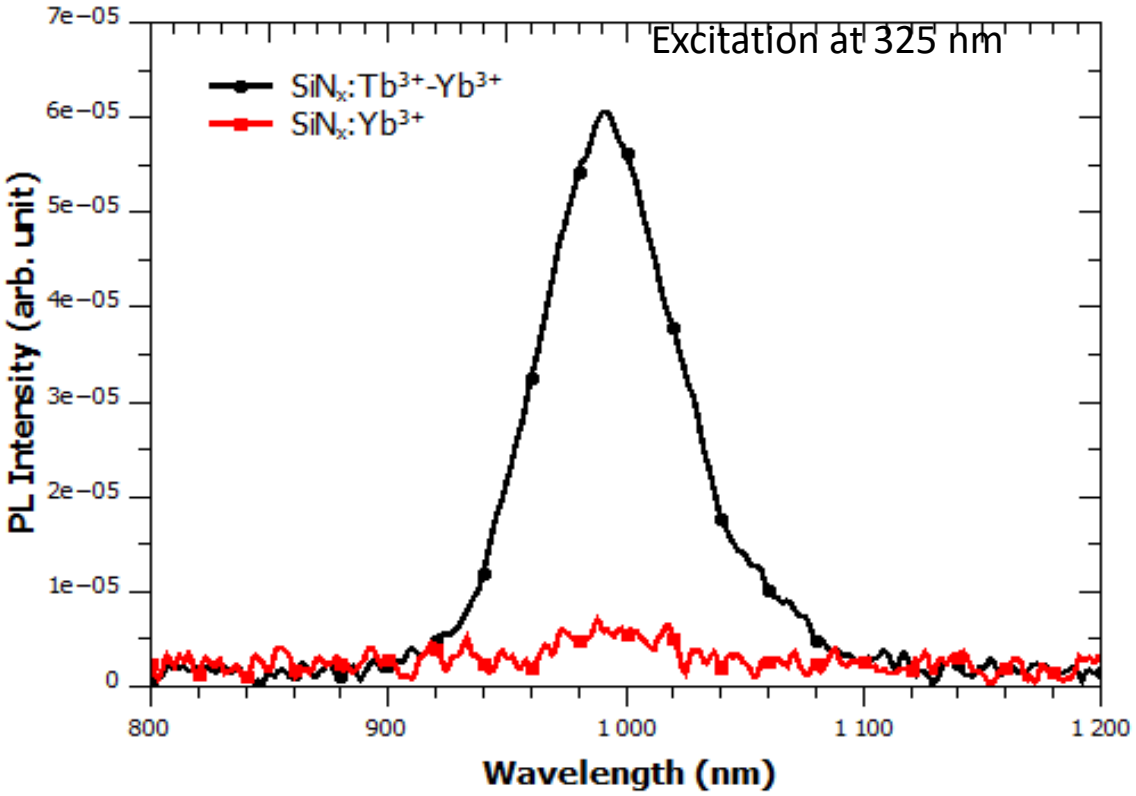




- Extinction of Tb^{3+} luminescence and Tb^{3+} lifetime decrease with [Tb] increase

→ Concentration quenching

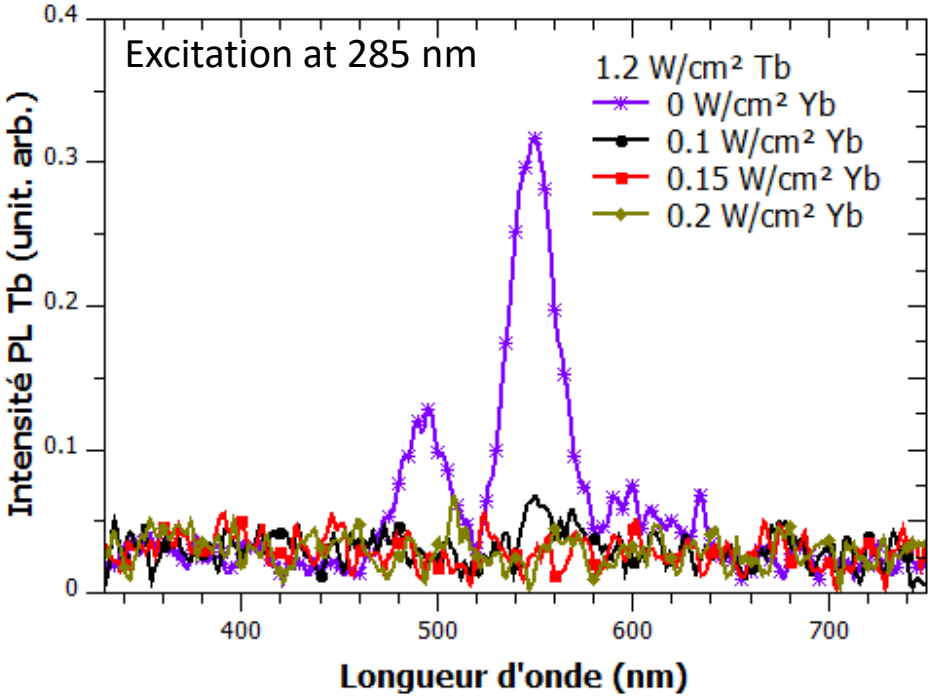
Energy transfer to Yb^{3+}

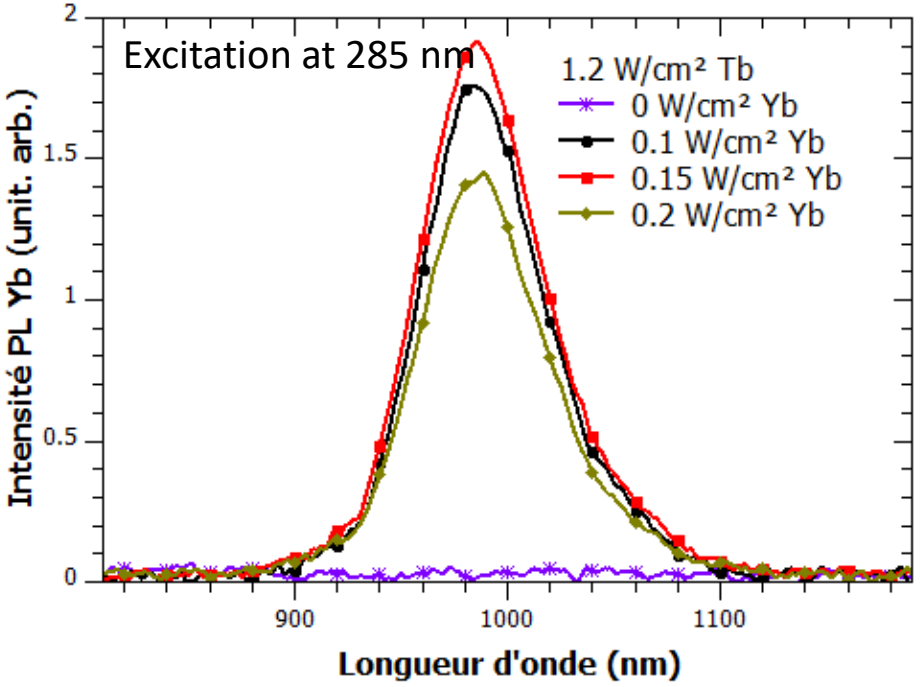


- Yb^{3+} PL Intensity appears in the presence of Tb^{3+} ions
- Non-resonant excitation of Yb^{3+}

→ Cooperative Energy transfer (CET) between Tb^{3+}d Yb^{3+} ions

Energy transfer from Tb^{3+} to Yb^{3+}





- Tb^{3+} PL Intensity decrease with increasing Yb^{3+} content
- Yb^{3+} PL Intensity increase with increasing concentration
- Non-resonant excitation of Yb^{3+}

→ Cooperative Energy transfer (CET) between Tb^{3+}d Yb^{3+} ions

$SiN_x:Tb^{3+}Yb^{3+}$

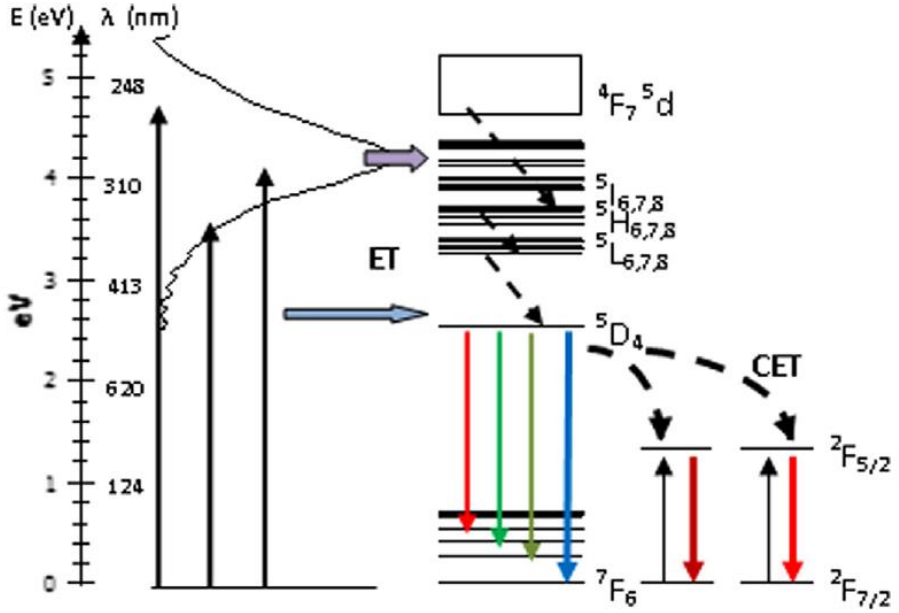


Fig. 5. Down-conversion process. A schematic of the energy levels of the down-conversion system with the excitation in black arrows, the radiative de-excitation (photon emission) in colored arrows, the non-radiative de-excitation in black straight dashed line arrows, the energy transfer in big blue arrows, and the cooperative energy transfer in black curved dash line arrows. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article)

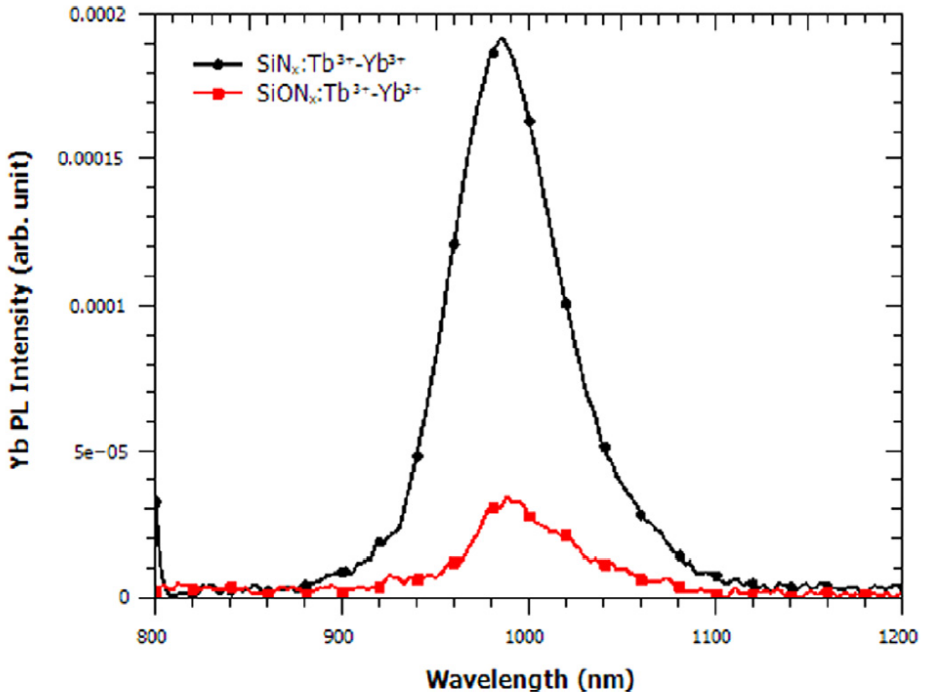
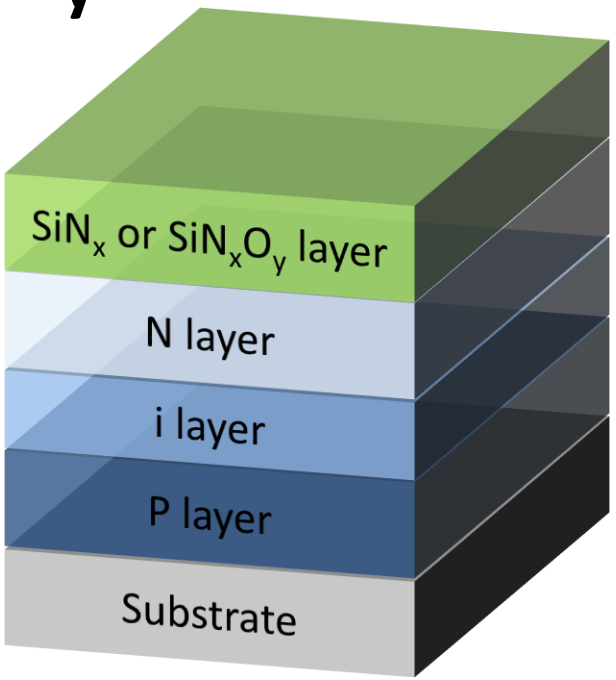


Fig. 9. Yb^{3+} emission for different matrixes. Photoluminescence spectra of the best samples obtained of SiN_x and SiO_xN_y co-doped matrixes for an excitation wavelength of 285 nm measured with a grating of 600 s/mm-750 bw.

$SiN_x:Tb^{3+},Yb^{3+}$ versus $SiN_xO_y:Tb^{3+},Yb^{3+}$

Table 1
Extraction efficiencies. Extraction efficiencies of the SiN_x and SiO_xN_y matrixes in the air, the layer, and the silicon.

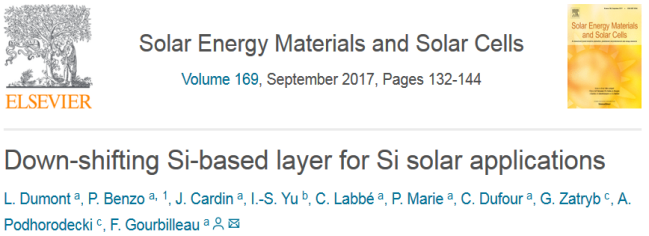
η^{ext} (%)	Air	Layer	Silicon
SiN_x	15.8	60.8	23.4
SiN_xO_y	22.8	59.7	17.5



- **Internal efficiency of SiN_x based film is about 29 times larger than the one obtained with SiN_xO_y based film**
- **External efficiency of SiN_x based film (with extraction into the Si solar cell) is about 12 times larger than the one obtained with SiN_xO_y based film**

- *Highly Efficient Infrared Quantum Cutting in Tb3+– Yb3+ Codoped Silicon Oxynitride for Solar Cell Applications, YT An et al, Advanced Optical Materials 1 (11), 855-862, 2013*
- *$SiN_x:Tb^{3+}–Yb^{3+}$, an efficient down-conversion layer compatible with a silicon solar cell process, L Dumont et al, Solar Energy Materials and Solar Cells, 2016*

Down shifting layer



Solar Energy Materials and Solar Cells
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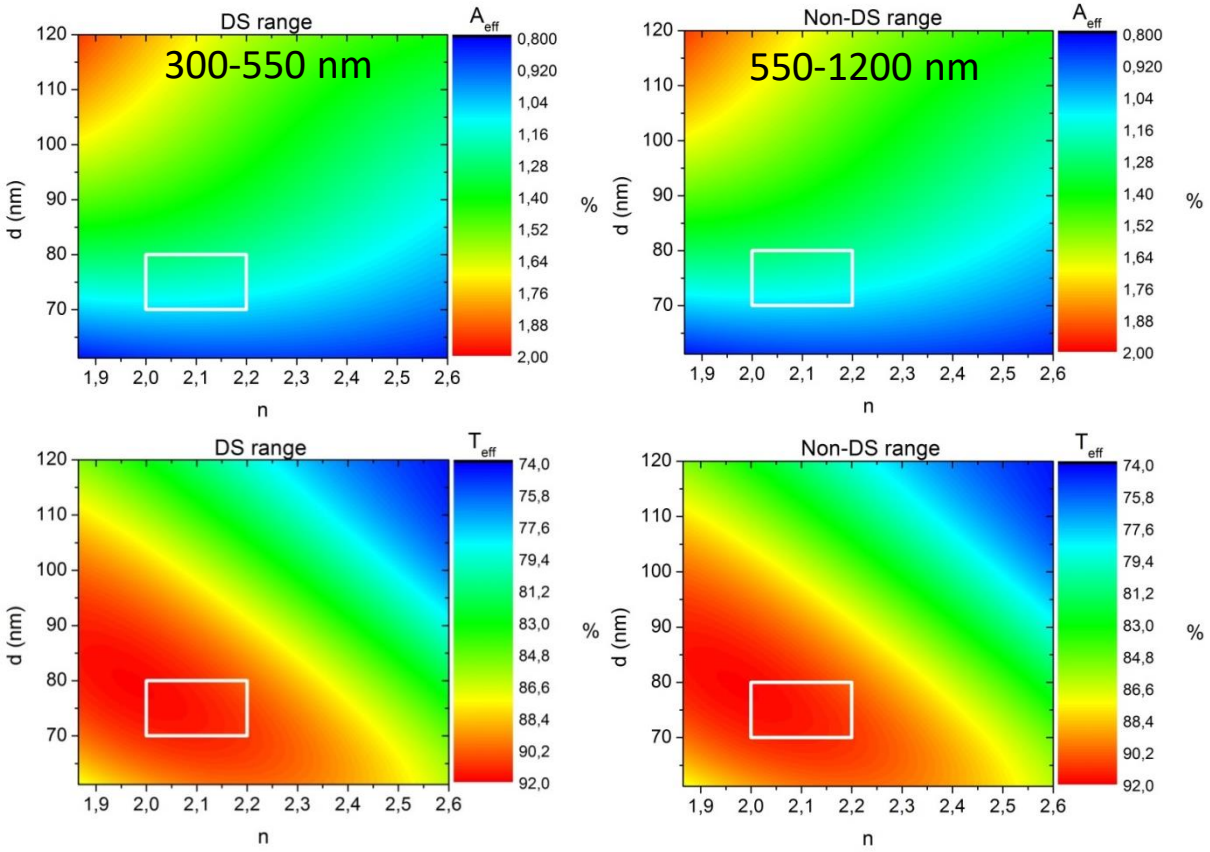
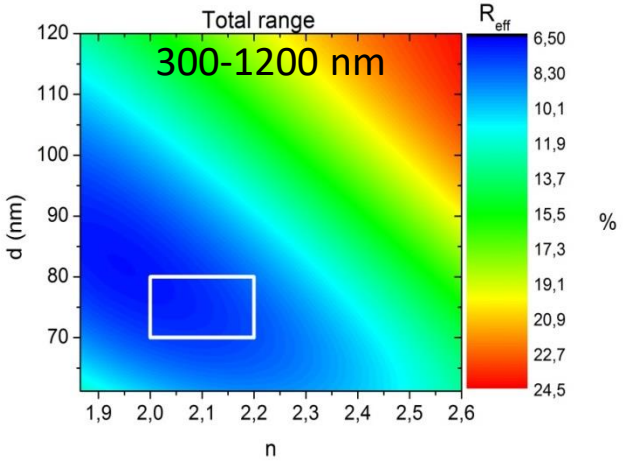
Down-shifting Si-based layer for Si solar applications

L. Dumont [a], P. Benzo [a, 1], J. Cardin [a], I.-S. Yu [b], C. Labbé [a], P. Marie [a], C. Dufour [a], G. Zatryb [c], A. Podhorodecki [c], F. Gourbilleau [a]

DS layer optical properties preliminary studies



$$A_{eff} = \frac{\int_{\lambda_{min}}^{\lambda_{max}} A(\lambda) I_{AM1.5}(\lambda) d\lambda}{\int_{\lambda_{min}}^{\lambda_{max}} I_{AM1.5}(\lambda) d\lambda}$$



- **A_{eff}** and **T_{eff}**

→compromise under n=2,2 and 100 nm

- **R_{eff}**

→2,0 < n < 2,2

→70 nm < d < 80 nm

PL Extraction efficiency $SiN_x:Tb^{3+}$

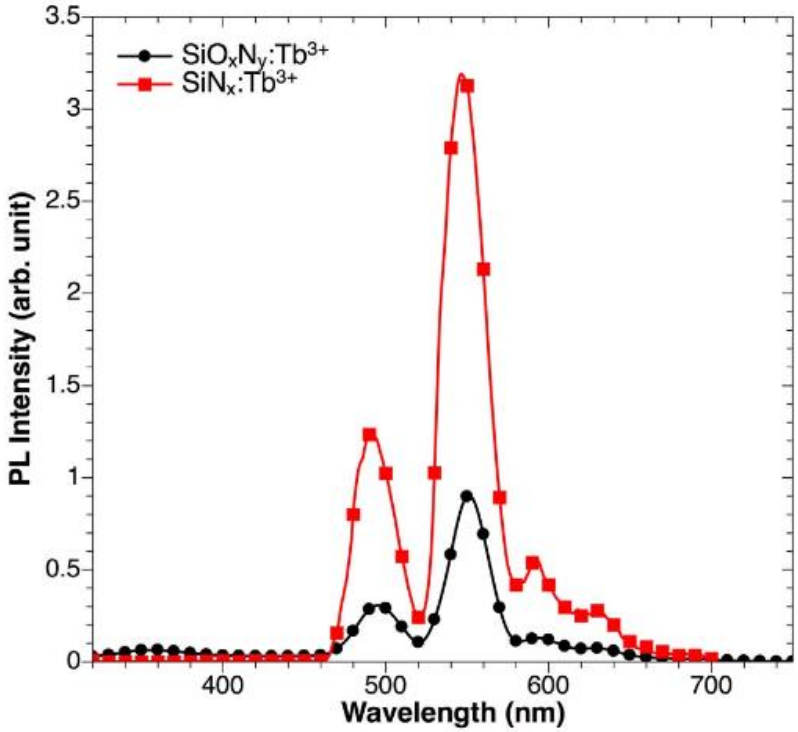


Fig. 15. Photoluminescence spectra of the optimized samples obtained of Tb^{3+}-doped SiO_xN_y and SiN_x doped terbium matrices for a 285 nm excitation wavelength measured with the 600 lines/mm-750 bw grating.

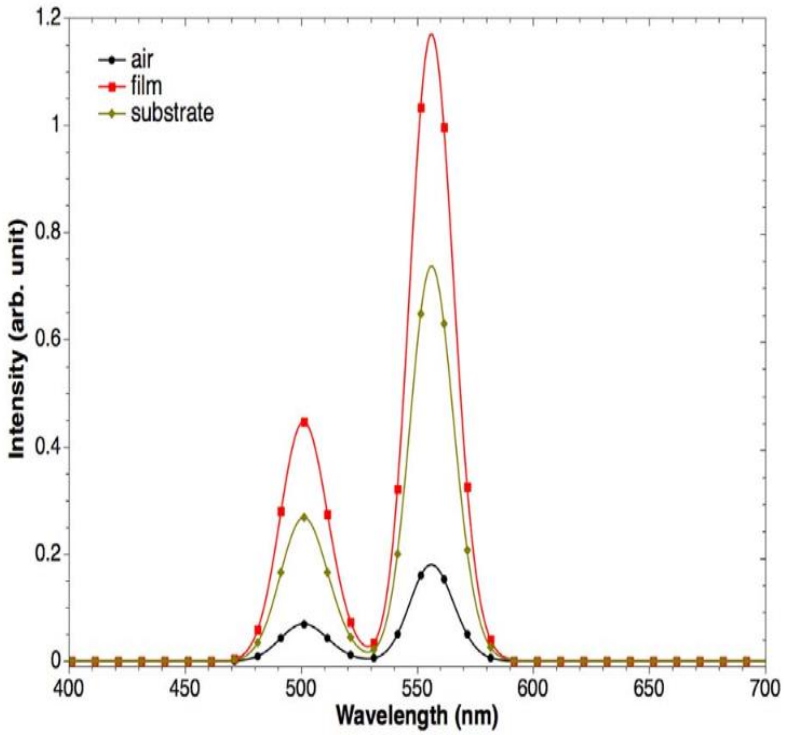


Fig. 1. Modeled Tb^{3+} *PL* spectra recorded in the three media, only the two more intense peaks/bands representing the $^5D_4$ to $^7F_6$ and $^5D_4$ to $^7F_5$ energy transitions are considered.

Table 2
Integrated luminescence efficiencies of the SiN_x and SiO_xN_y matrixes in the air, the layer, and the silicon.

η^{lum} (%)	Air	Layer	Silicon
SiN_x	8.7	56.3	35
SiN_xO_y	13.8	60.2	26

Larger efficiency of the DS layer with the SiN_x matrix than with SiN_xO_y due to larger Tb content and to larger extraction into the Si PV cell

- *Down-shifting Si-based layer for Si solar applications, L Dumont et al, Solar Energy Materials and Solar Cells 169, 132–144, 2017*

Integration in an industrial process

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RESEARCH ARTICLE

WILEY PROGRESS IN PHOTOVOLTAICS

First down converter multilayers integration in an industrial Si solar cell process

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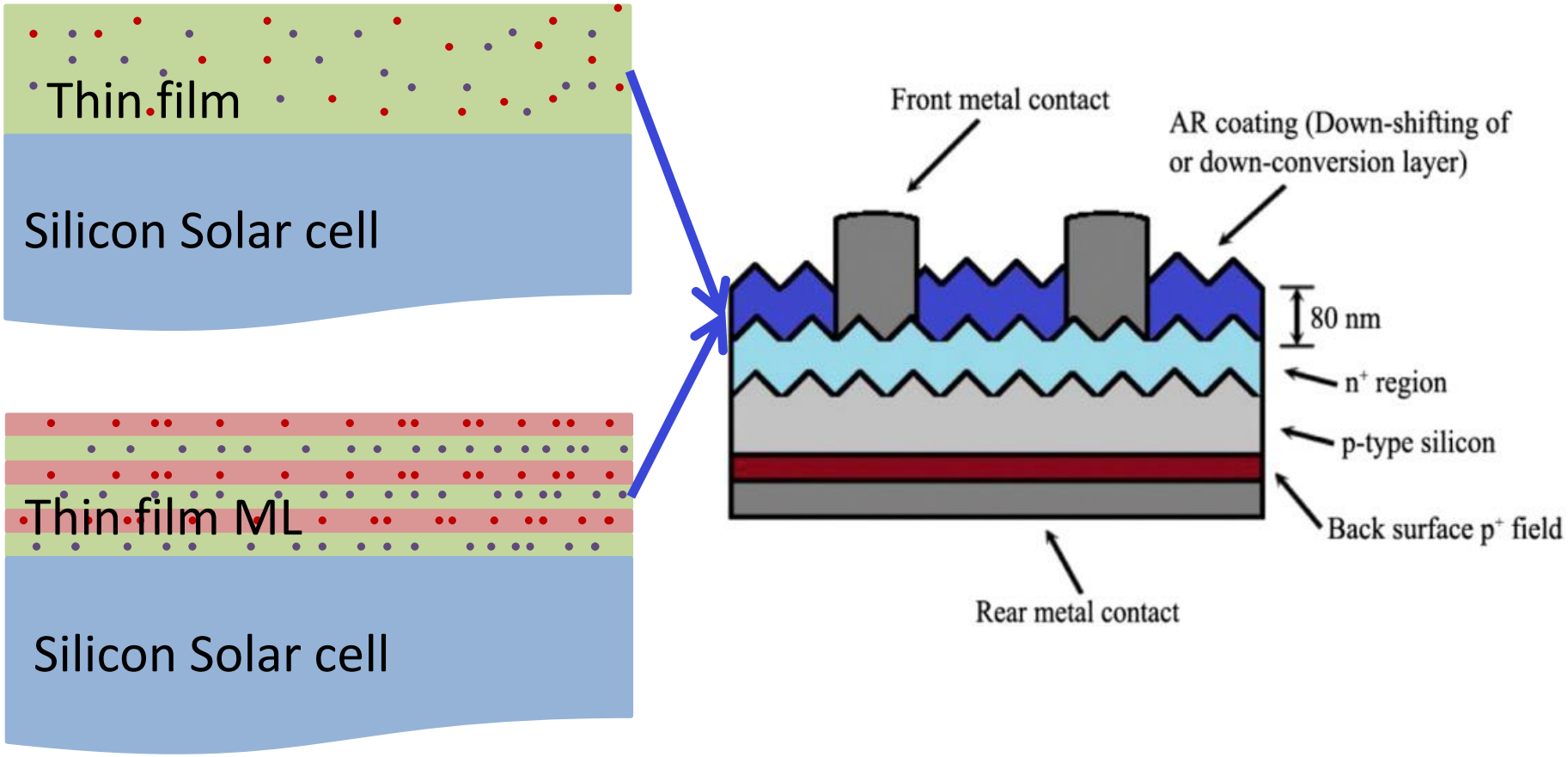
ELSEVIER

Journal of Rare Earths
Available online 7 November 2018
In Press, Accepted Manuscript

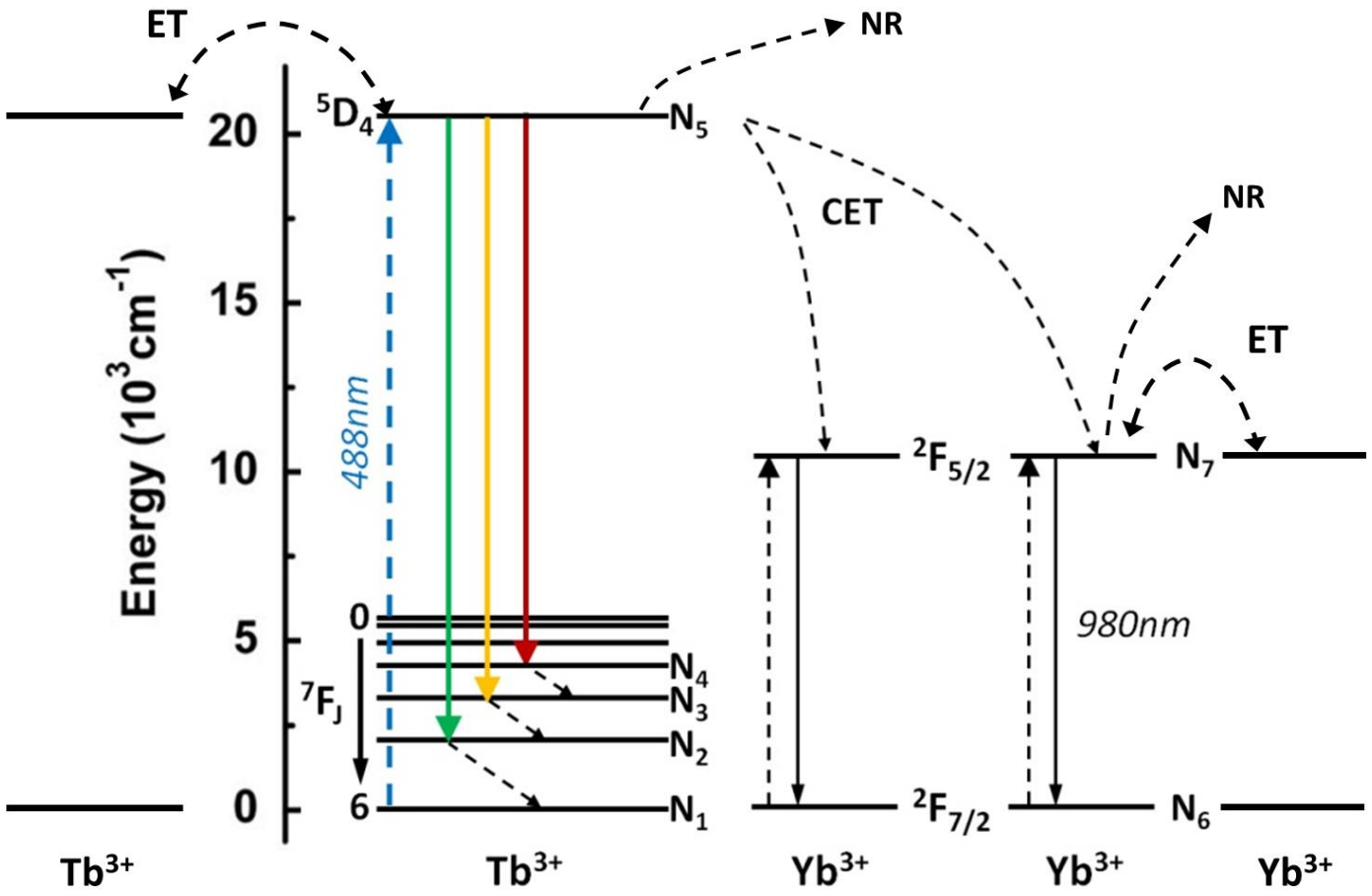
Monolithic crystalline silicon solar cells with SiN_x layers doped with Tb^{3+} and Yb^{3+} rare-earth ions ☆

Integration in an industrial process



Modeling of Donor (Tb^{3+})-Acceptor(Yb^{3+}) ions system

Modeling of Donor(Tb^{3+})-Acceptor(Yb^{3+}) ions system



Donor(Tb^{3+})-Acceptor(Yb^{3+}) ions system

Rate equations modeling of steady state of electronic energy levels

$$\frac{dN_1}{dt} = -\sigma\phi\,(N_1 - N_5) + \frac{N_5}{\tau_{51}^r} + \frac{N_2}{\tau_{21}^{nr}} + K_{CET}N_5N_6^2 + \frac{N_5}{\tau_{51}^{nr}} + k_{ET}(Tb^{3+})N_5$$

$$\frac{dN_2}{dt} = \frac{N_5}{\tau_{52}^r} + \frac{N_3}{\tau_{32}^{nr}} - \frac{N_2}{\tau_{21}^{nr}}$$

$$\frac{dN_3}{dt} = \frac{N_5}{\tau_{53}^r} + \frac{N_4}{\tau_{43}^{nr}} - \frac{N_3}{\tau_{32}^{nr}}$$

$$\frac{dN_4}{dt} = \frac{N_5}{\tau_{54}^r} - \frac{N_4}{\tau_{43}^{nr}}$$

$$\frac{dN_5}{dt} = \sigma\phi\,(N_1 - N_5) - \sum_{j=1}^{4}\frac{N_5}{\tau_{5j}^r} - K_{CET}N_5N_6^2 - \frac{N_5}{\tau_{51}^{nr}} - k_{ET}(Tb^{3+})N_5$$

$$\frac{dN_6}{dt} = -2K_{CET}N_5N_6^2 + \frac{N_7}{\tau_{76}^r} + \frac{N_7}{\tau_{76}^{nr}} + k_{ET}(Yb^{3+})N_7$$

$$\frac{dN_7}{dt} = 2K_{CET}N_5N_6^2 - \frac{N_7}{\tau_{76}^r} - \frac{N_7}{\tau_{76}^{nr}} - k_{ET}(Yb^{3+})N_7$$

Cooperative energy transfer between Tb^{3+} Yb^{3+} ions

Energy transfer between Tb^{3+} ions (cross-relaxation)

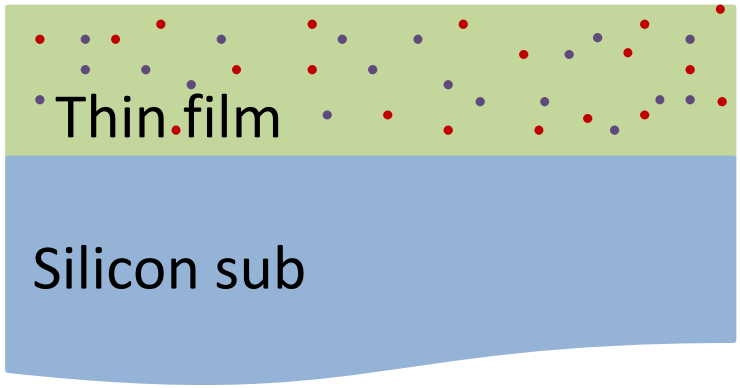
energy transfer between Yb^{3+} ions (cross-relaxation)

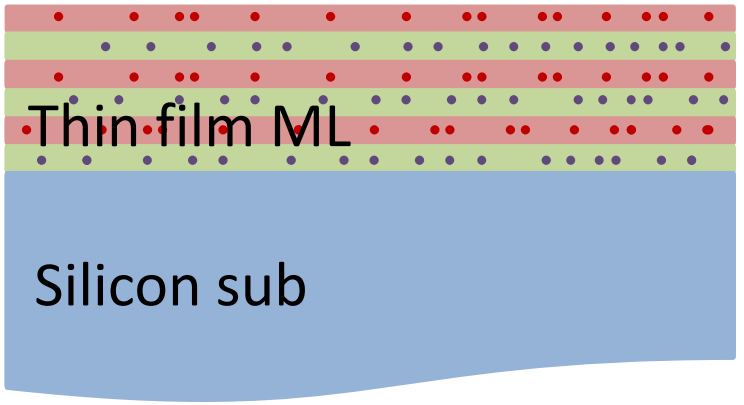
Look on modelling parameters

- K_{CET}, $K_{ET}(Tb^{3+})$ and $K_{ET}(Yb^{3+})$ distance dependent (r) and therefore function of concentration $[Tb^{3+}]$ and $[Yb^{3+}]$

$$K_{CET} \propto \frac{1}{r^{6}_{Tb^{3+}-Yb^{3+}} \cdot r'^{6}_{Tb^{3+}-Yb^{3+}}} \qquad K_{ET}(RE^{3+}) \propto \frac{1}{r^{6}_{RE^{3+}-RE^{3+}}}$$

- Those parameters are depending on statistical distribution of rare earth ion in thin films



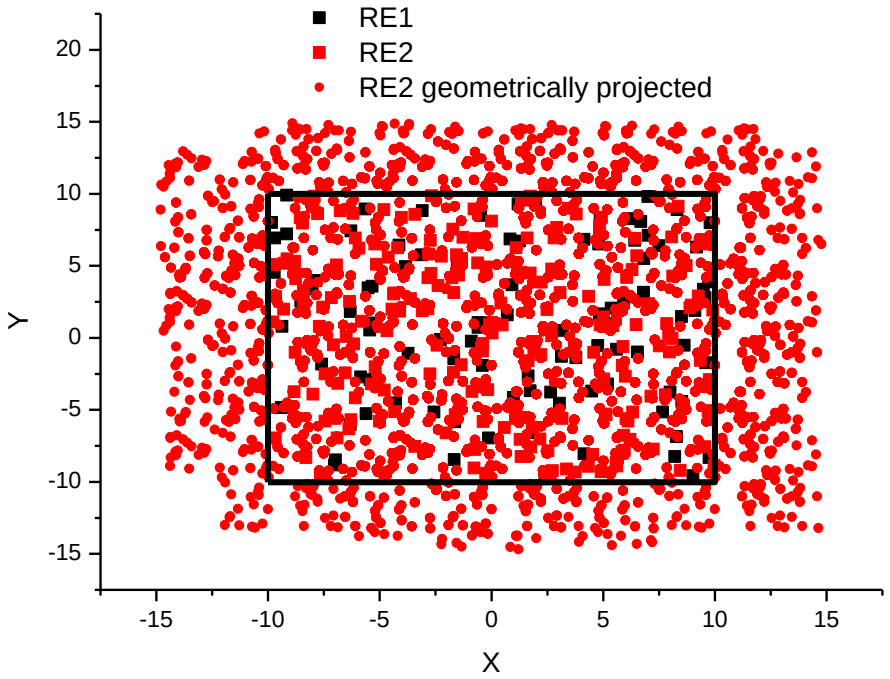


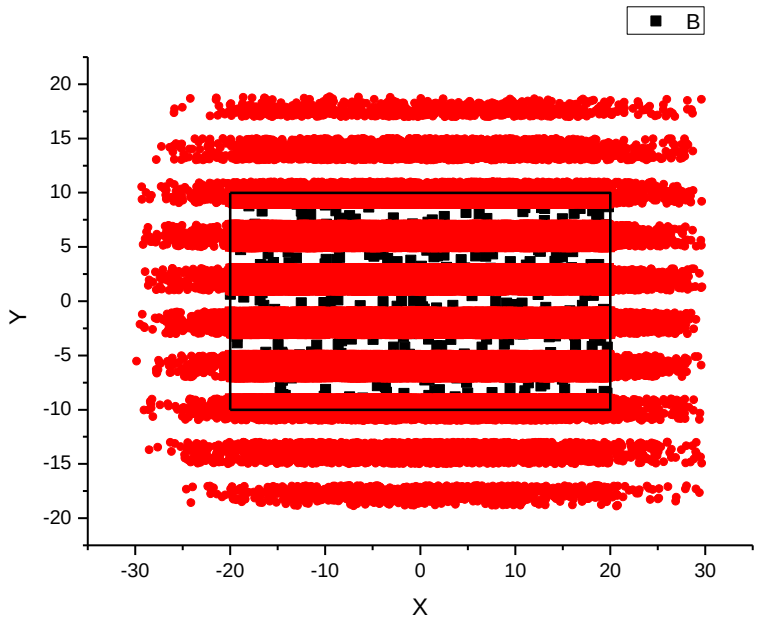
- Homogenous thin film and ML thin film allow to reach different kind of spatial distribution and concentration of rare earths ions

$[RE^{3+}] \leq 10^{+20}$ cm^{-3} $[RE^{3+}] \leq 10^{+21\text{-}22}$ cm^{-3}

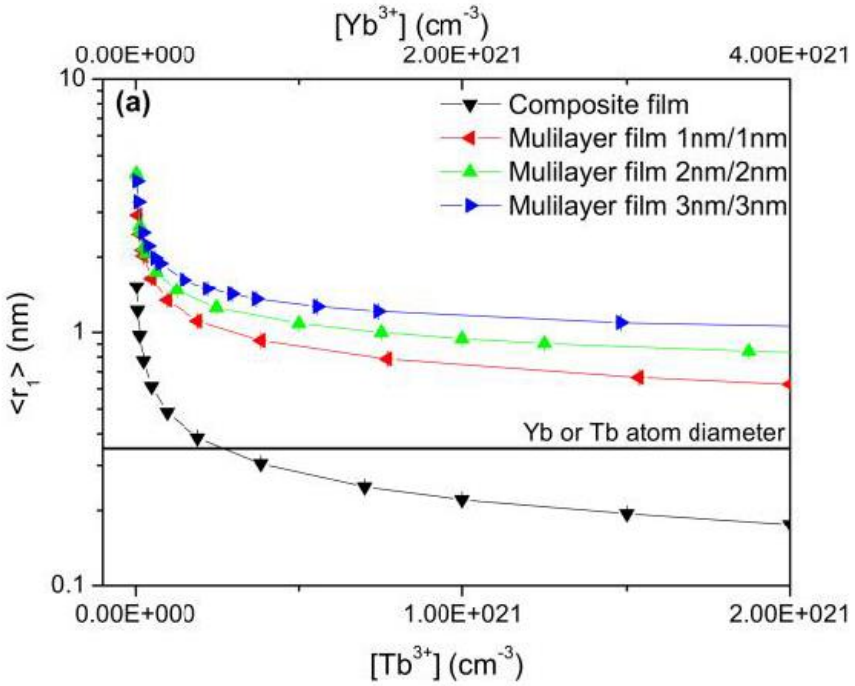
Comparison of homogenous thin film and multilayer systems

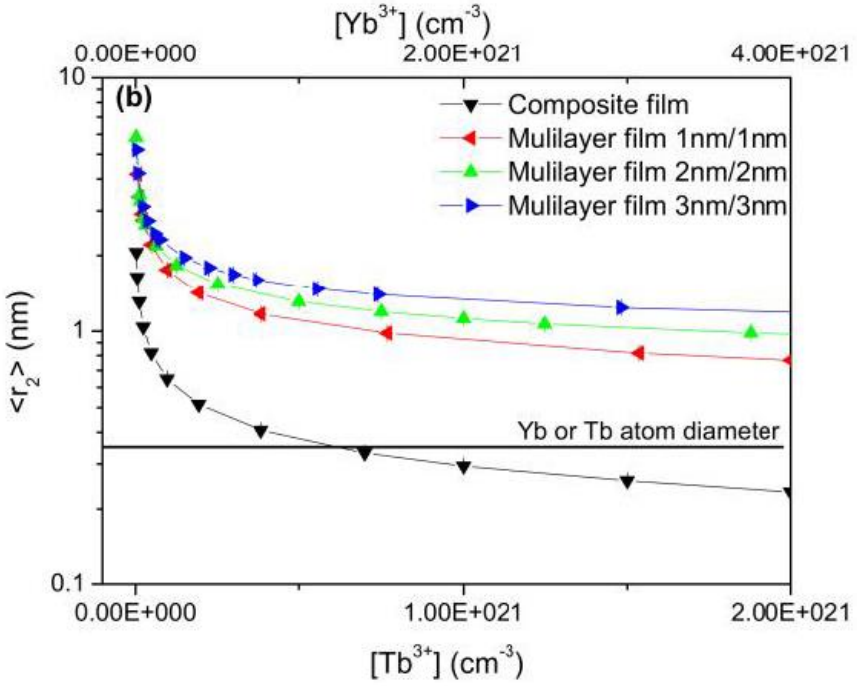
- Monte Carlo method for determination of average distance between first, second…neighbors





Thin film and multilayer systems

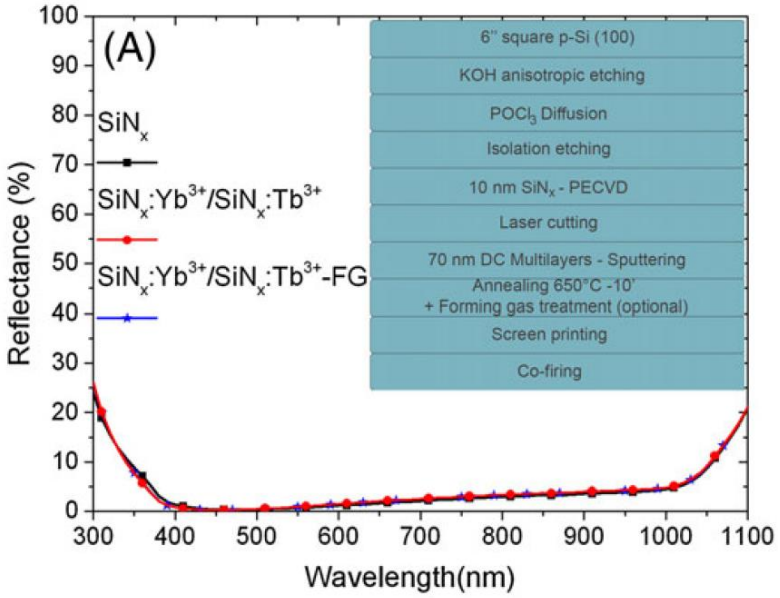




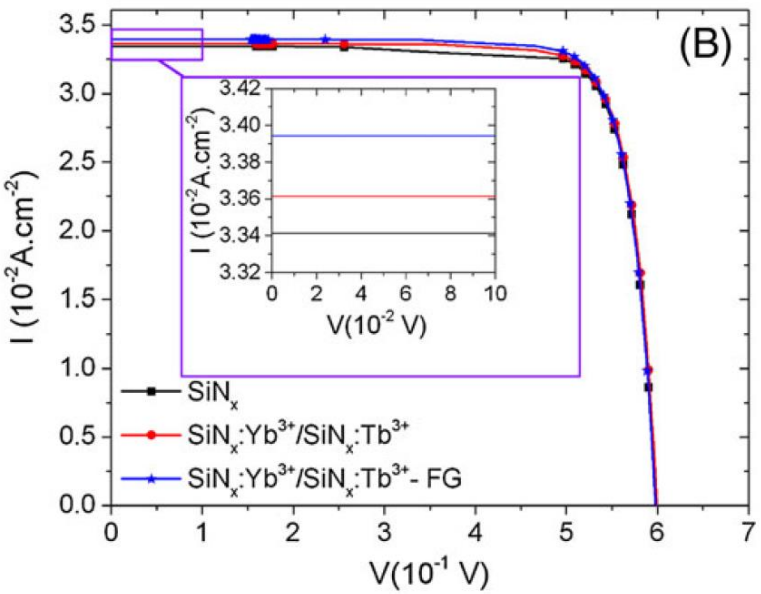
→ ML prevent aggregation quenching and allow therefore higher doping rate

Integration in an industrial process

Multilayer Integration on silicon solar cell



Reflectance of three layers (SiNx, SiNx:Yb3+/SiNx:Tb3+ *MLs*, SiNx:Yb3+/SiNx:Tb3+ *MLs* with forming gas post treatment) deposited on textured emitters.



I-V curves corrected of series resistance for three c-Si solar cells topped with SiNx, SiNx:Yb3+/SiNx:Tb3+ *MLs* and SiNx: Yb3+/SiNx:Tb3+ *MLs* with forming gas post treatment

ARC Layer	V_{oc}, V	J_{sc}, mA·cm^{-2}	Pseudo FF	Pseudo Efficiency (%) ± 0.03%
SiN_x	0.605	33.41	0.82	16.48
XC25(1.5/1.5)	0.599	33.61	0.82	16.53
XC25(1.5/1.5) + FG	0.598	33.94	0.82	16.66

Established Improvement of solar cell efficiency

Monolayer Integration on silicon solar cell

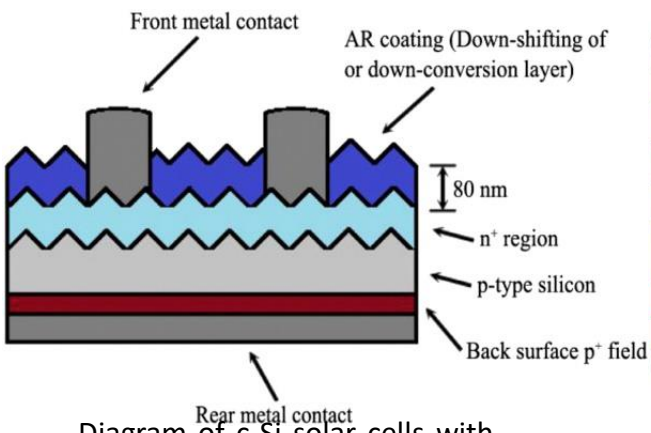
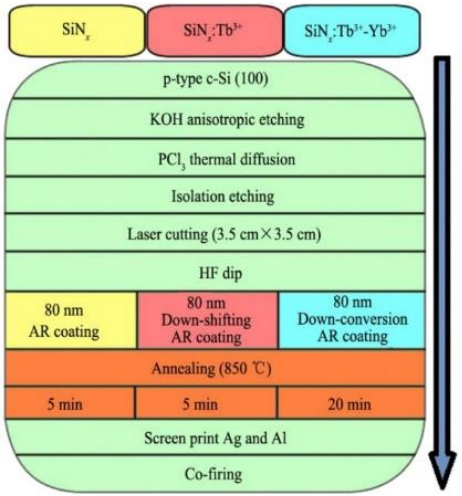
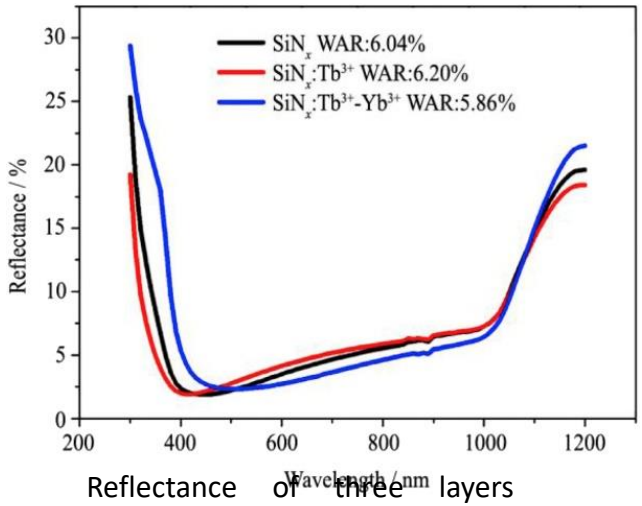


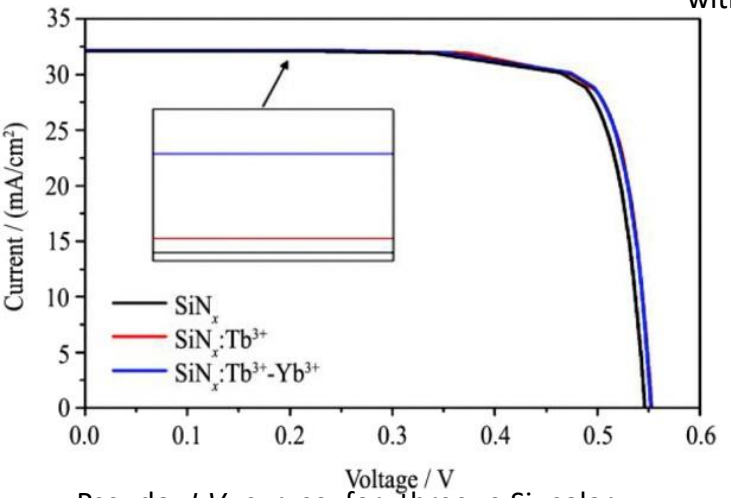
Diagram of c-Si solar cells with spectrum conversion layers



Process flow of c-Si solar cells with spectrum conversion layers.



Reflectance of three layers deposited on textured emitters



Pseudo *I-V* curves for three c-Si solar cells with SiN_x, SiN_x:Tb^{3+} and SiN_x:Tb^{3+}-Yb^{3+} layers, respectively.

Samples	J_{sc} (mA/cm^2)	V_{oc} (V)	Pseudo FF (%)	Pseudo Efficiency (%)	Efficiency to normalized transmission (%)
SiN_x	32.091	0.546	0.804	17.13	18.23
SiN_x:Tb^{3+}	32.098	0.552	0.803	17.31	18.45
SiN_x:Tb^{3+}-Yb^{3+}	32.165	0.552	0.806	17.36	18.44

PV performances of c-Si solar cells with SiN_x, SiN_x:Tb^{3+} and SiN_x:Tb^{3+}-Yb^{3+} layers

relative increase by 1.34% in the conversion efficiency

Conclusion

- Established results:
  - SiN_xO_y and SiN_x based DC, DS layer
  - Integration of ML and homogenous DC layer in an industrial process of Si PV cell
  - **relative increase by 1.34% in the conversion efficiency**
- Perspectives:
  - Ce^{3+} emission in SiN_xO_y/SiN_x layer ->increase of excitability
  - Distance distribution model in material -> further improvement
  - Up conversion layer with Er^{3+}

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