Energy Materials

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Outline

1. Criticality in Materials

2. Criticality in Energy Materials

Two examples of research in
energy-related material science

3. How to replace a material:
   Transparent Conductor Oxides

4. How to modify material properties:
   Quantum confinement effects in Ge Nanostructures

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The production, conversion, distribution and use of Energy critically depends on Materials!

…what beyond this obvious statement?

1) Supply of existing materials?

2) New materials?

3) New technologies?

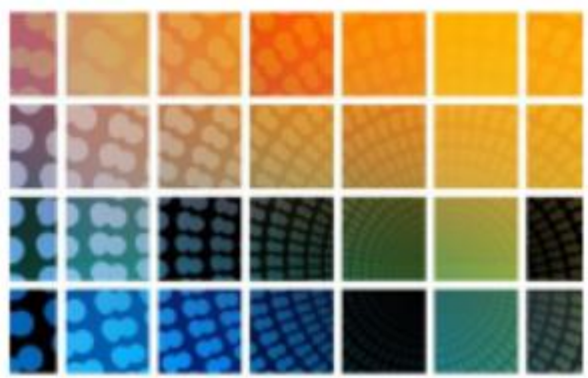
4) …other?

There is a strong concern and care about the Energy-Materials Nexus

REPORT ON

Forward Looking Workshop on Materials for Emerging Energy Technologies

Edited by
Dr. Johan Veiga Benesch



EUROPEAN COMMISSION

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Directorate G – Industrial Technologies
Unit G3 – Materials
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…something I did not expect!

3.2 Bamboo Composites for Wind Power

The role of bamboo in the development of environmentally friendly wind turbines.

Photo 3.2.1: Example of 5-layered, vertical strips, bamboo plywood

Forward Looking Workshop on Materials for Emerging Energy Technologies

The conclusions of the experts include that:

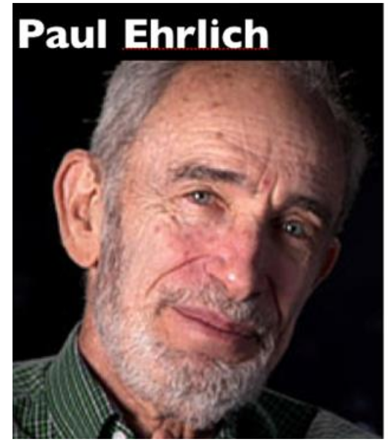
- No single technology would be a total solution in itself and that developing an energy mix will become even more important in the future.
- Europe must take the lead in development of energy technologies and associated technologies.
- An understanding of technology fundamentals, up-scaling and tech demonstrators are all very important in the development of new energy technologies.
- There is an opportunity to promote research that aims at finding synergies of a wide mix of energy technologies.

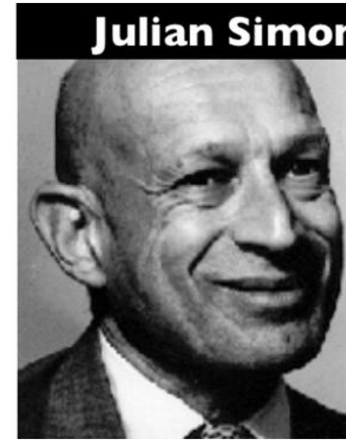
The experts' recommendations on actions identified needs for:

- Further research into the structure and properties of materials for energy.
- Further research into new materials or materials solutions.
- Sufficient funding to overcome the obstacles in the up-scaling of the emerging energy technologies.
- There is a need for much more multi-disciplinary activity to take place.
- Advanced computer based complexity modelling
- Recognition there are no simple fixes for power generation in the 2050 horizon.

Can we predict the trend?

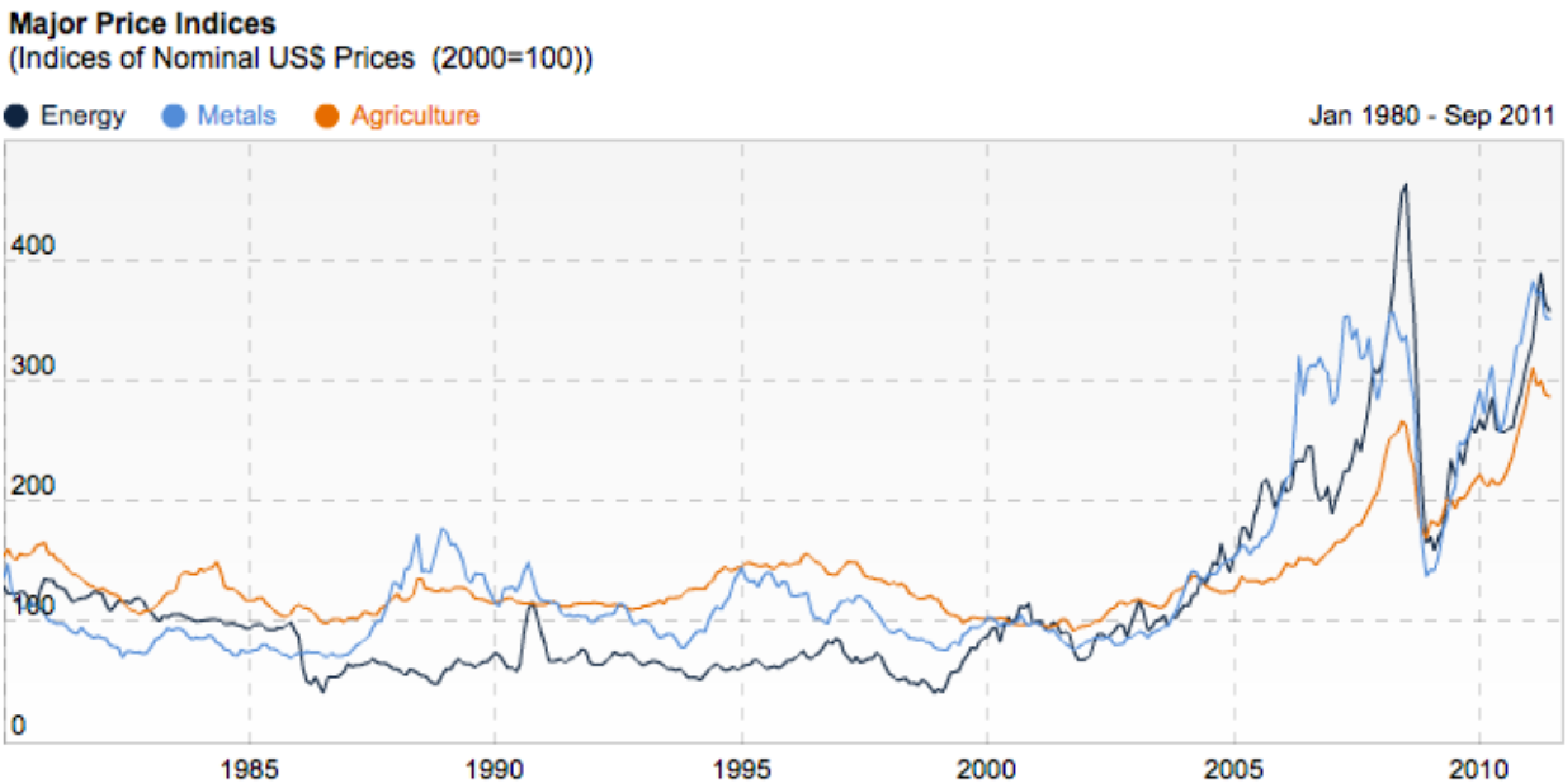
The Ehrlich-Simon Bet
1980





- Paul Ehrlich (professor of Population Studies at Stanford University)
  - Metal prices will be higher in 10 years because of ever-increasing demand driven by relentless population growth.
- Julian Simon (professor of Business Administration at the University of Maryland)
  - Prices will be lower because technology will make the extraction of the metals cheaper, or we will find alternatives for those that are really running out.

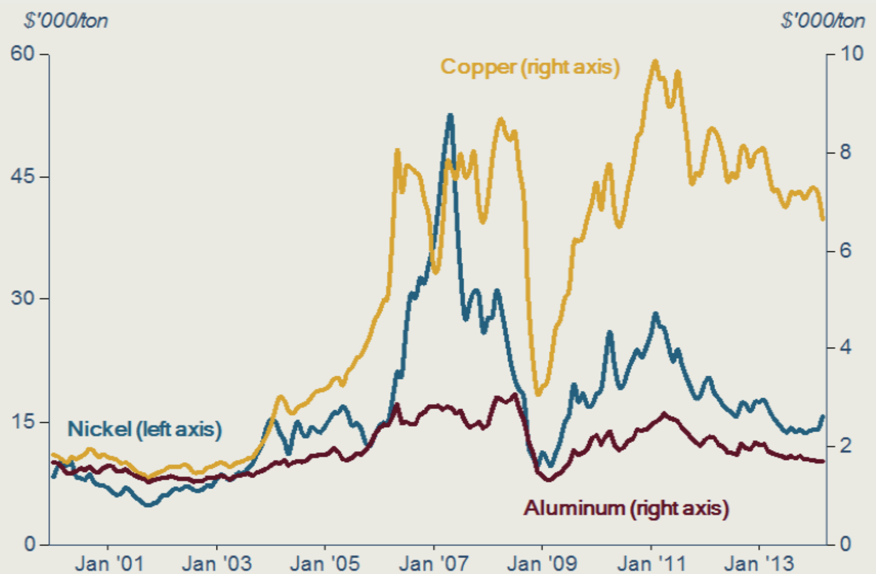
Is there a "hockey-stick" effect? Sudden fluctuations!



Source: World Bank

Real criticality or policy/market instability?

Figure 12 **Aluminum, copper and nickel prices**



Source: World Bank.

Figure 13 **Lead, tin, and zinc prices**

Source: World Bank.

Tony Terrasi: “Energy Materials”, EPS-SIF Joint School on Energy, Varenna (Italy), July 21-26 2017

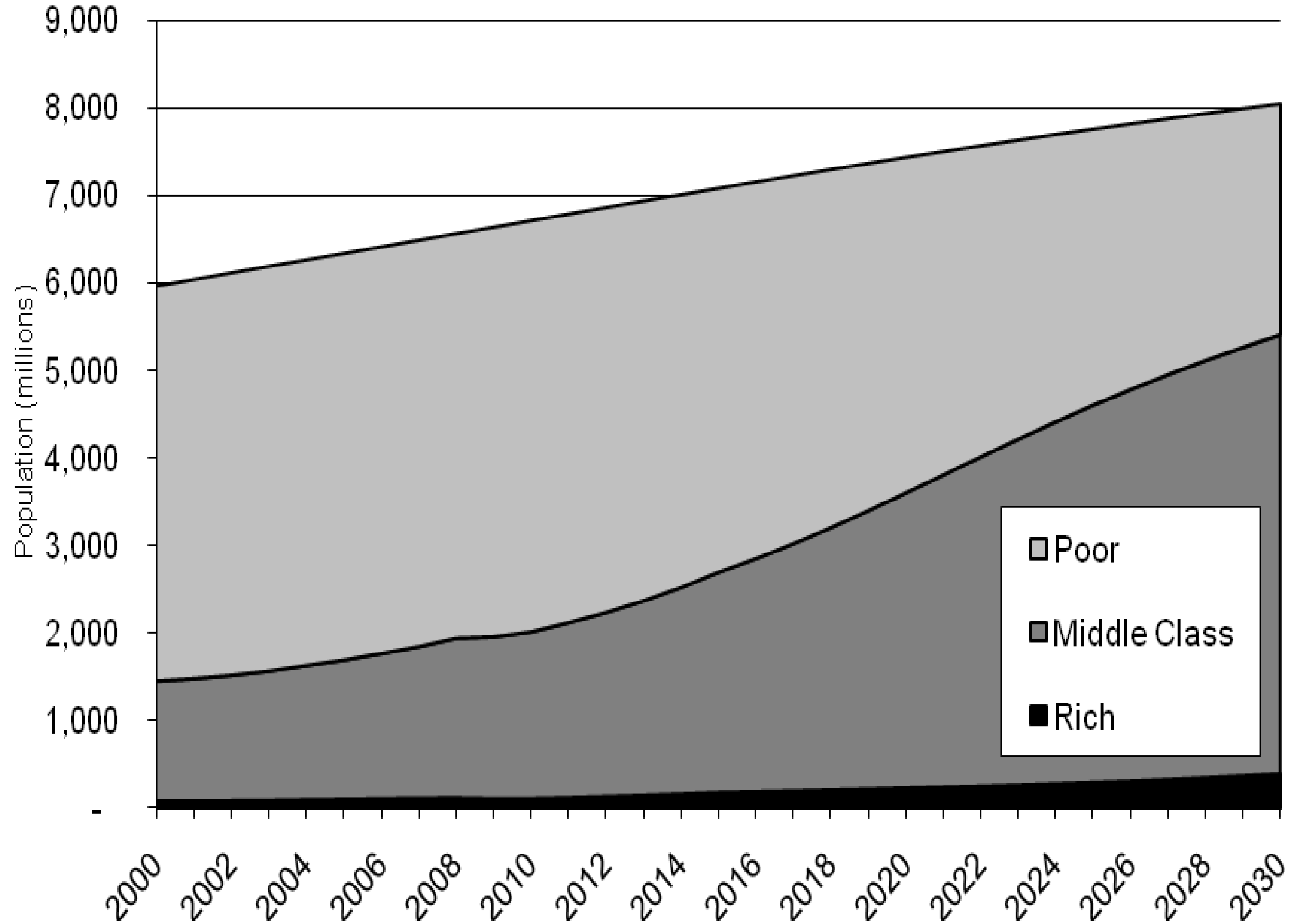
Factors influencing the criticality

Middle class growth

OECD (Organization for Economic Co-operation and Development) expects the world's middle class to grow -

from1.8 billion people, in 2012,

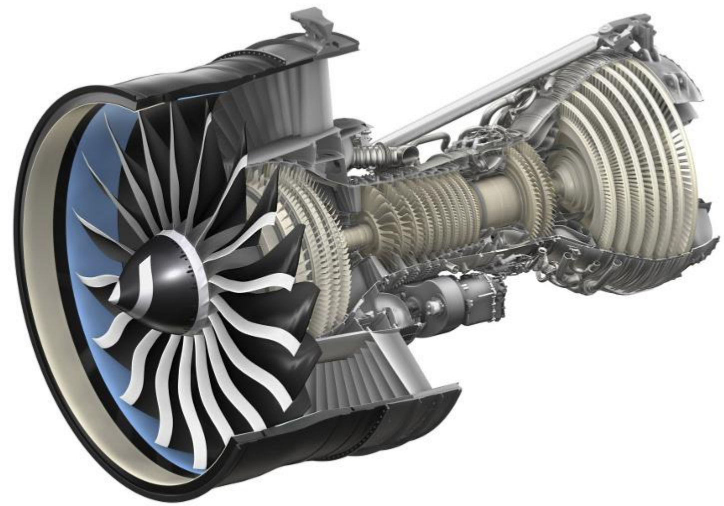
to 4.9 billion in 2030.



Source: Wolfensohn Center for Development, at Brookings

Some examples of materials criticality affecting us *today*

- Jet engine manufacturers, including CMI partner GE, have had to deal with shortages of rhenium (Ni-based alloys). Re is by-product of Cu and Mo.



- A major disk-drive manufacturer came within one week of shutting down production for lack of Nd-Fe-B magnets.

Some examples of materials criticality affecting us *today*

- Loudspeaker manufacturers have been severely impacted by magnet price increases (rare earths Nd and Sm-Co).

- Tesla Motors may be forced to reduce production because of short supplies of Li-ion batteries.

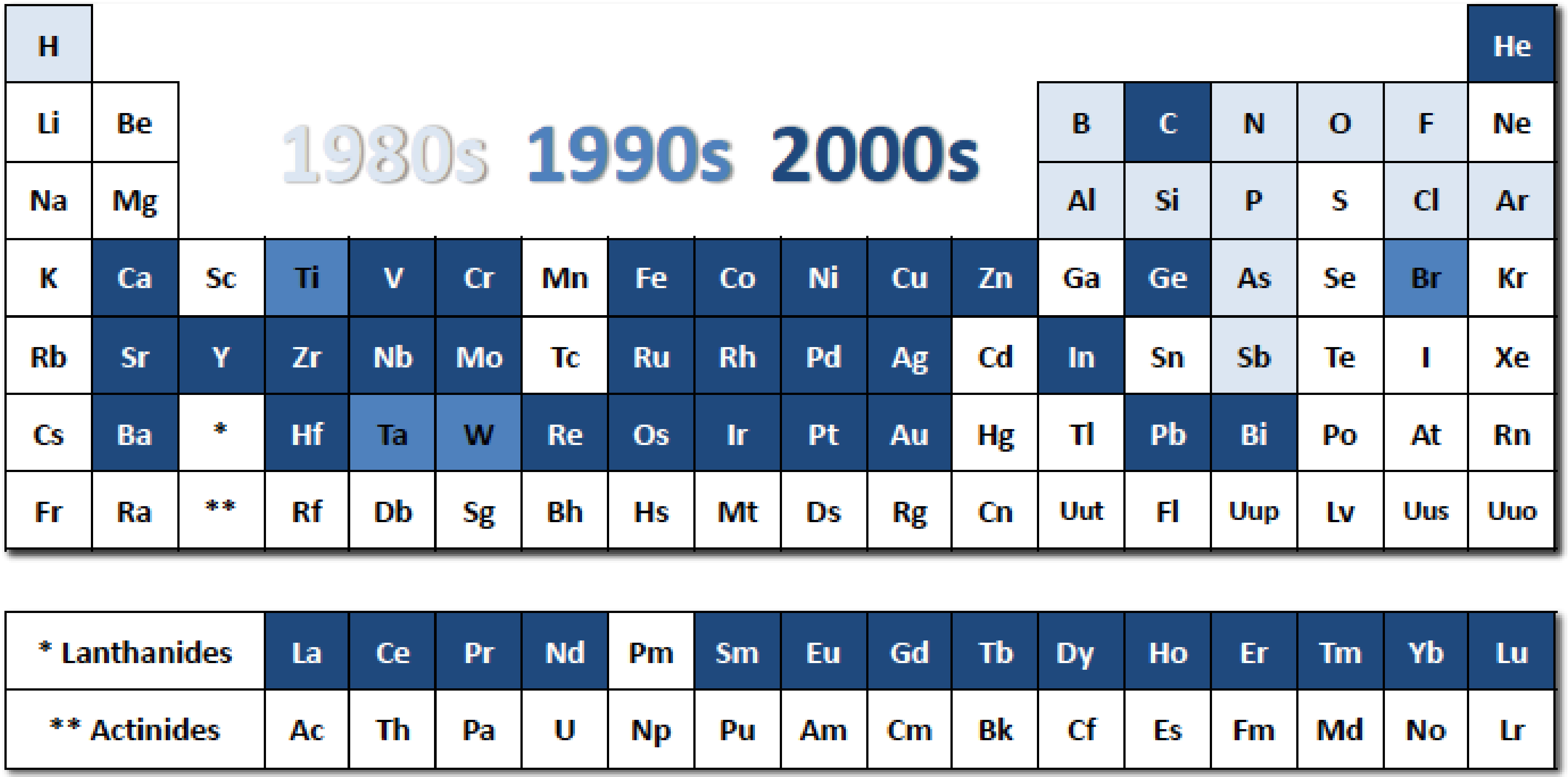
Some examples of materials criticality affecting us *today*

- The target date for transition to high-output T5 fluorescent lamps has been delayed by two years because manufacturers claim that there is a shortage of Eu and Tb for the phosphors.

- Utility-scale wind turbine installations are overwhelmingly gearbox-driven units, despite the high failure-rate of the gearboxes, because of the cost and unavailability of Nd and Dy required for direct-drive units.

More functions, more elements

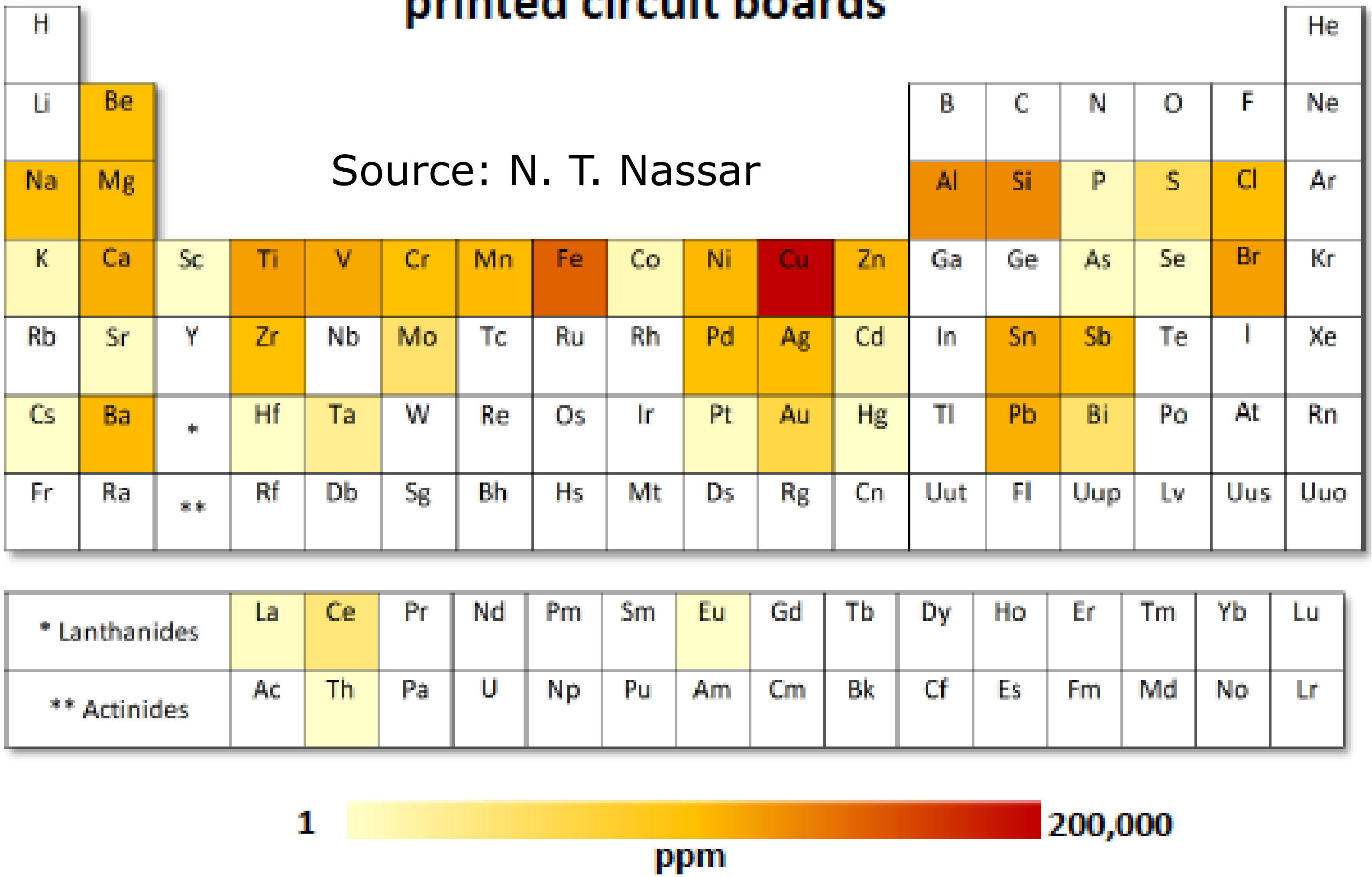
Elements utilized in computer chip manufacturing over three decades



Source: N.T. Nassar

Adapted from: T. McManus, Intel Corp., 2006

Concentration of 44 elements found in printed circuit boards



Data source : UNEP (2013) Metal Recycling: Opportunities, Limits, Infrastructure, A Report of the Working Group on the Global Metal Flows to the International Resource Panel. Reuter, M. A.; Hudson, C.; van Schaik, A.; Heiskanen, K.; Meskers, C.; Hagelüken, C.

- Complexity begets vulnerability -

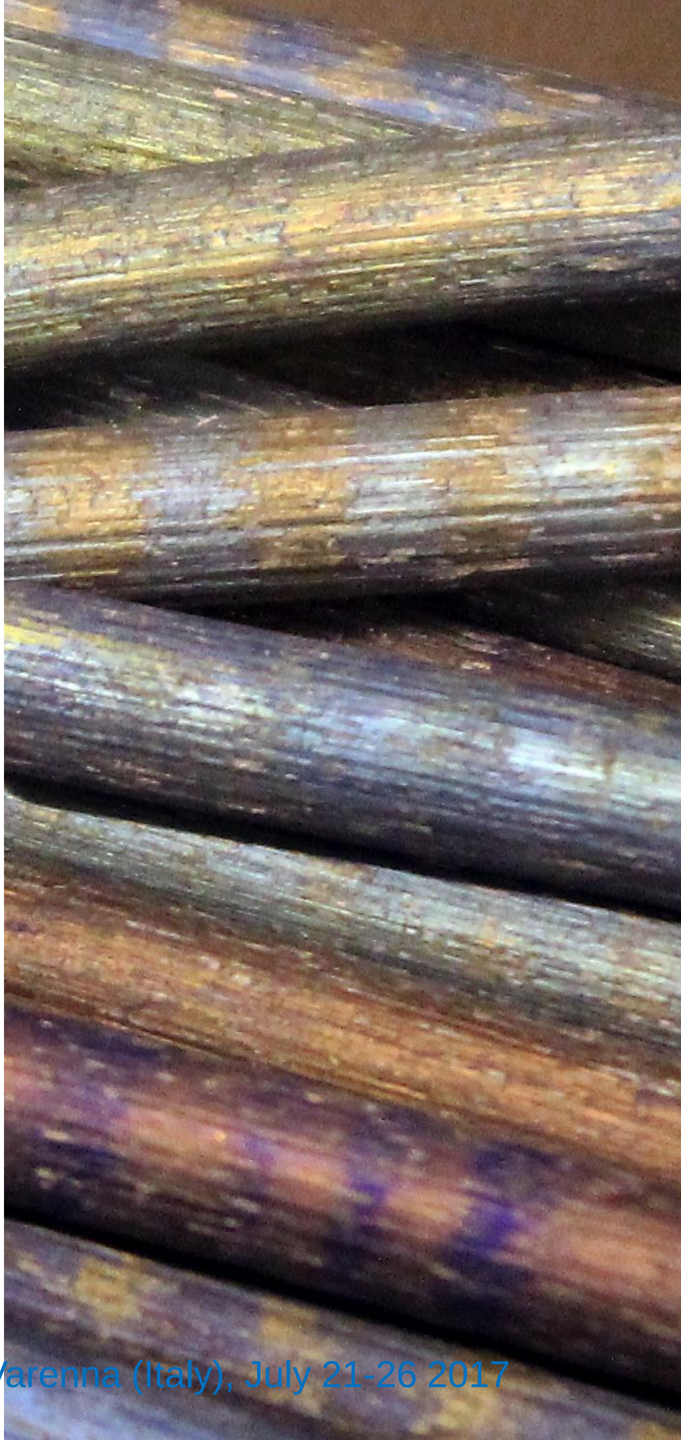
~30 elements

~75 elements

What is a "Critical Material?"

- Any substance used in technology that is subject to supply risks, and for which there are no easy substitutes or, in plain English, stuff you really need but can't always get.

- The list of materials that are considered critical depends on *who*, *where* and *when* you ask.



Critical Materials are not new: learning from the history

- “The stone age did not end because we ran out of stones” – Steven Chu.

- The copper age replaced the stone age because copper was better for some things.

- The bronze age replaced the copper age because bronze was better than copper.

- But the bronze age was not replaced by the iron age because iron was better than bronze. It ended because copper became unavailable.

Iron vs. Bronze, 1200 BC

- Processing
  - Bronze requires lower temperatures
- Hardness
  - Bronze is better, because no effective hardening mechanisms are yet available for iron.
- Corrosion
  - Bronze is better
- Cost
  - Iron was nine times more expensive than gold

The Bronze Age Collapse

~1200 BC

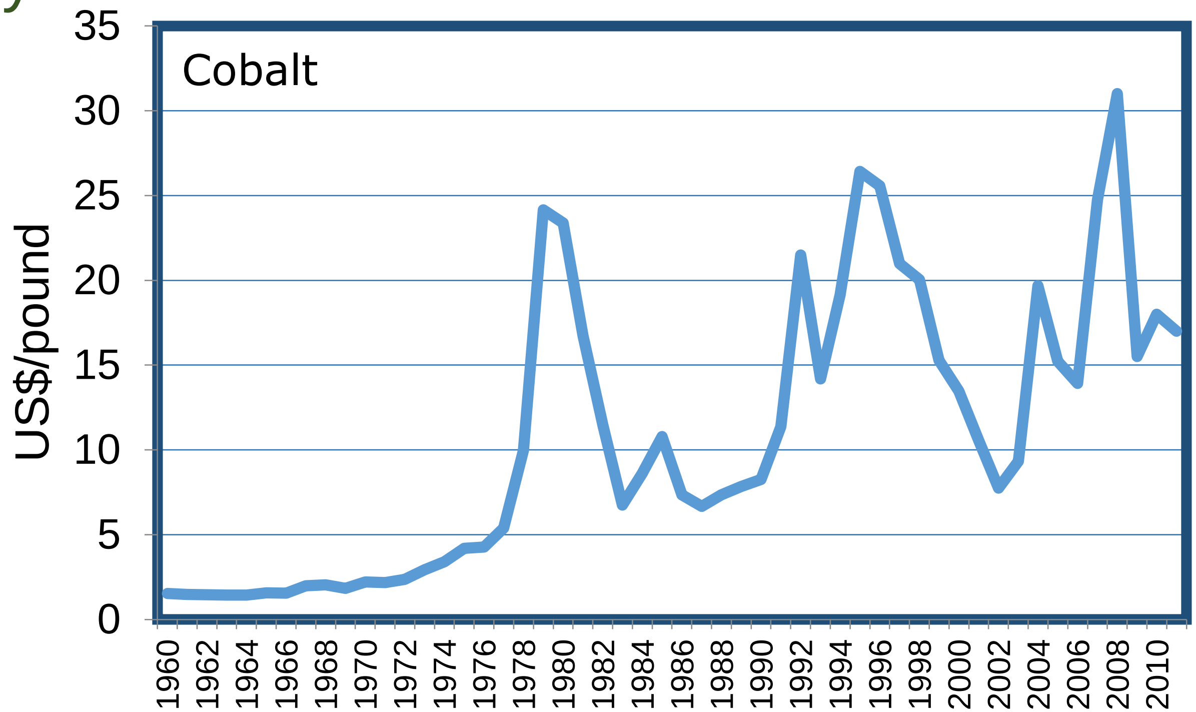
- Bronze becomes unavailable
  - Possibly because Cyprus is overtaken by war, making copper inaccessible.
- Results
  - Collapse of trade; collapse of civilization
  - Relative strengthening of Egypt, which found alternative sources in Africa
  - Eventual emergence of the iron age
- Responses include
  - Recycling
  - Source Diversification
  - Materials Substitution

We are working on the problems of today to prepare for the problems of tomorrow

- Better foresight through improved economic analysis
- Faster response through improved technical capabilities
- Learning from industry
- Learning from history

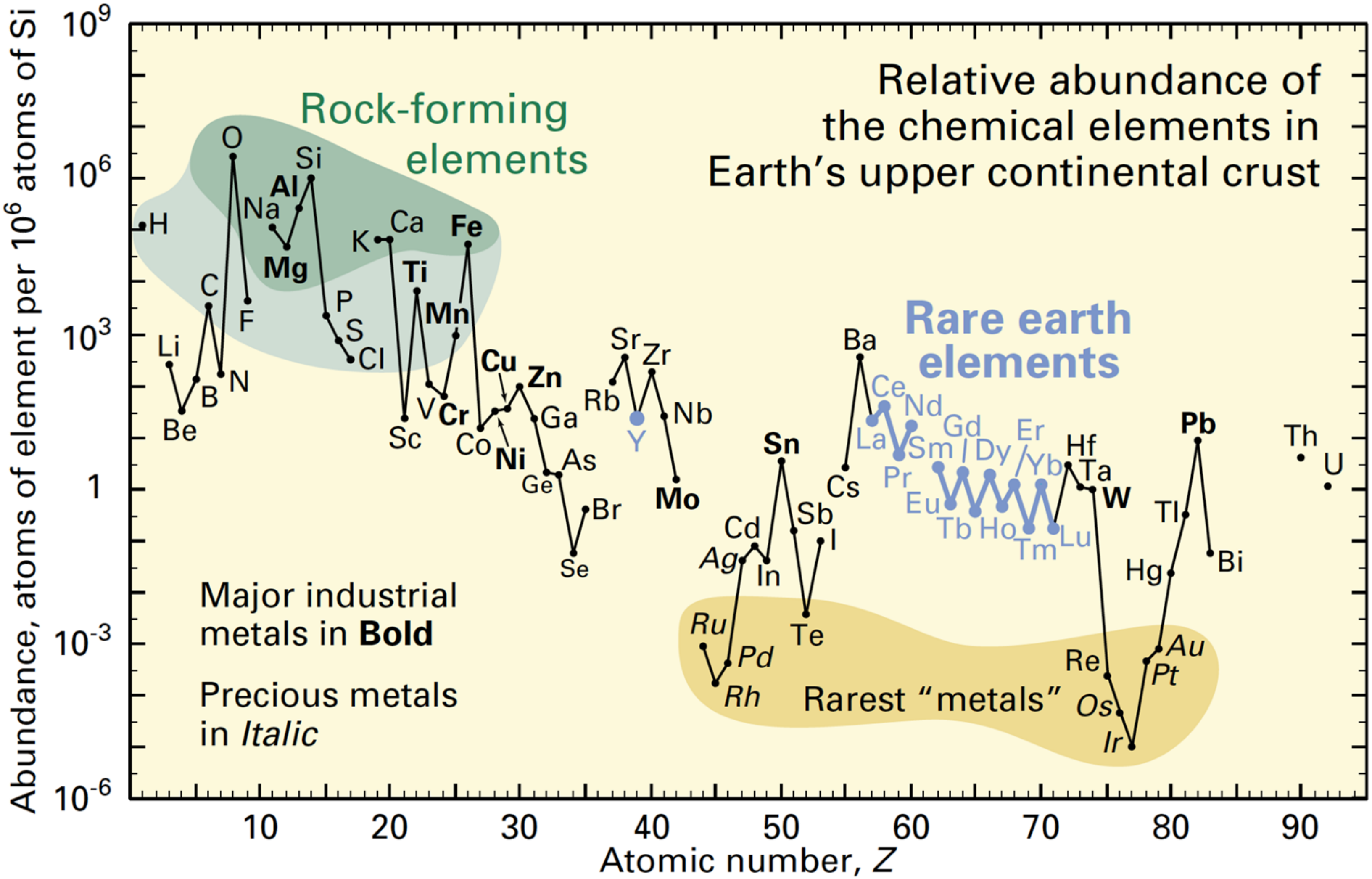
Criticality is a recurring phenomenon, and there is reason to believe that it will increase in frequency in the coming decades.

History shows that criticalities do not tend to have smooth recoveries.



The Rare Earths case

"Rare Earths" are neither rare, nor earths



The didymium nexus

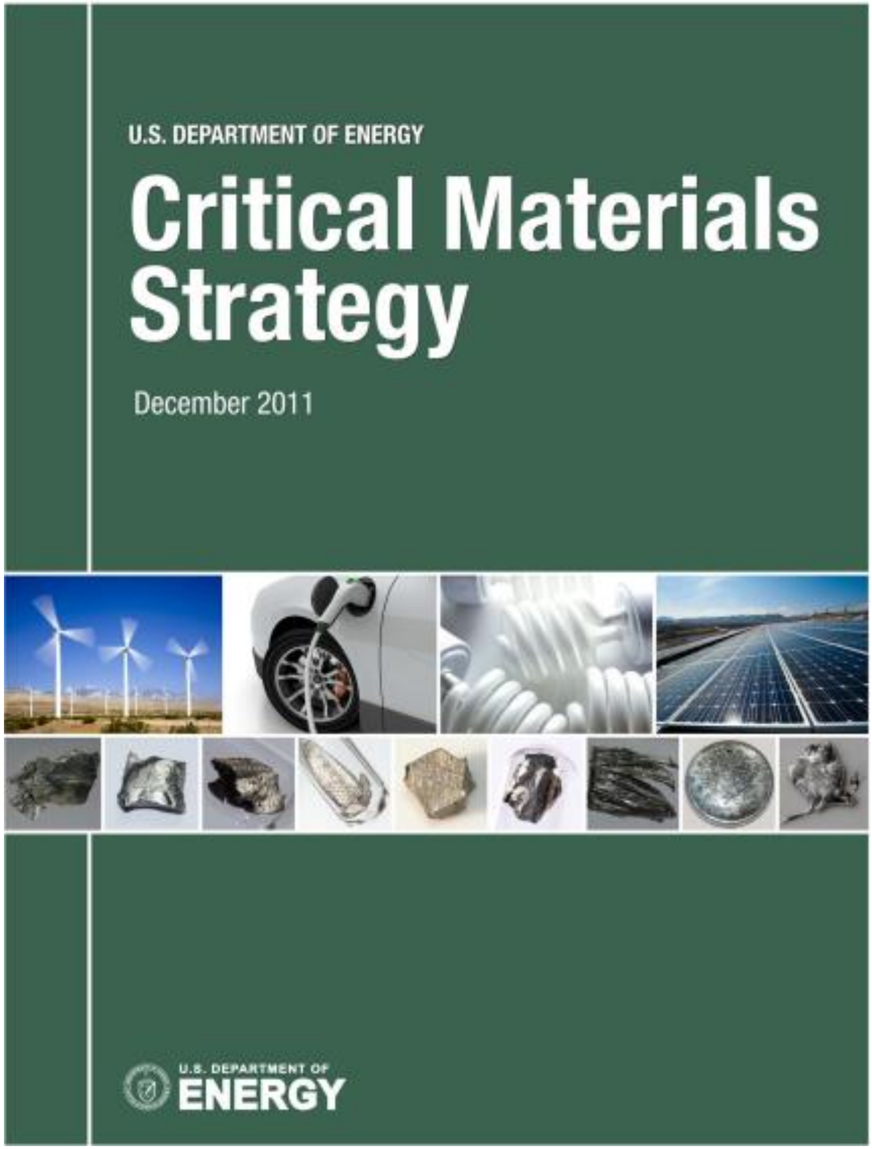
21 Sc														
39 Y														
57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu

- Praseodymium and neodymium are adjacent “light” rare earths.
- They are difficult to separate, and often sold un-separated, as “didymium,” which works well in many rare earth magnets.
- New uses for *Pr* are emerging:
  - Dopants in ZnO varistors
  - Replacing toxic yellow pigments for ceramics and polymers.
- *Pr* prices and *Nd* prices have decoupled and *Pr* is now ~30% more expensive.
- Less *Pr* use in magnets drives up demand for *Nd*, and adds separation costs.

Three-D Approach

- Diversify supply
- Develop substitutes
- Drive reuse, recycling, and efficient use of materials in manufacturing

Some of these approaches work better than others for specific materials.



Unfortunately, these approaches are slow to have impact

- Mine development, *where there is a known resource*, takes about 15 years, and has costs in the billions of dollars.

- Development and deployment of new materials takes an average of 18 years.

- There are no empirical data to suggest how long it takes for recycling programs to have an impact.

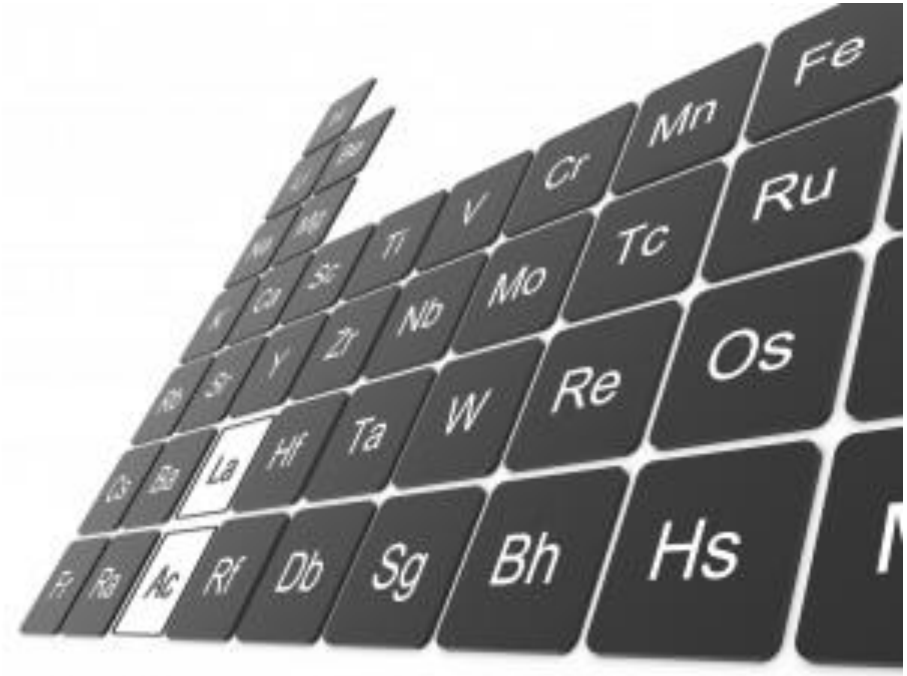
Two universal grand challenges

- Starting sooner
  - We need to anticipate criticality, not just respond to it.

- Working faster
  - 200 years at ~1000 BC
  - 20 years at ~ 2000 AD
  - 2 years by ~ 5000 AD?

Emergence of new critical materials

- Supply shortfalls
- Demand spikes
- Can we predict?
- Are we prepared to respond?



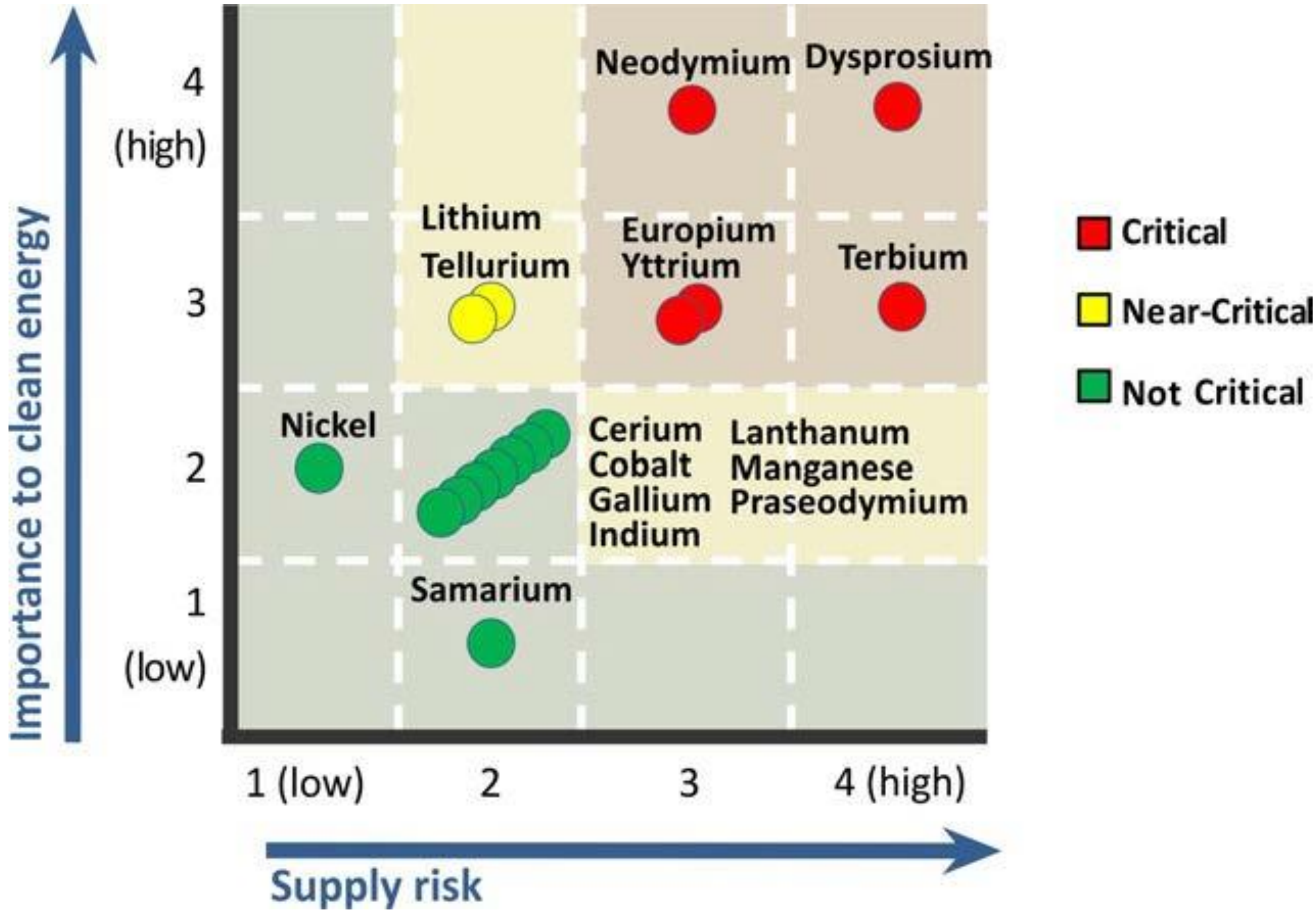
What is 'critical' for energy?

Many energy technologies, many possible *critical* materials . . .

- **Batteries**
  - Lead acid
  - NiMH: La, Ni
  - Lithium ion: Co, Li, graphite
  - Emerging: V, Mn
- **Solar**
  - Si based: Ag, Ni, Sn
  - CIGS: In, Ga, Se
  - CdTe
- **Magnets**
  - Ferrite
  - Alnico
  - SmCo
  - NdFeB: Nd, Pr, Dy, Tb
- **Lighting**
  - Fluorescent: Ce, Eu, La, Mn, Tb, Y
  - LED: Ce, Eu, Ga, Ge, In, La, Ni, Ag, Tb, Sn, Y
- **Catalysts**
  - Pt, Pd, Rh, Ce, La
- **Fuel cells**
  - Pt, Pd, Rh, La, Co, Ce, Y, Gd
- **Nuclear**
  - Co, Gd, Hf, In
- **Hydrogen electrolysis**
  - Pt, Pd, Rh

U.S. Department of Energy, medium-term assessment, 2015-2025

Source: U.S. Department of Energy (2011)



Joint Research Centre, European Commission (2013)

- 17 technologies, 33 elements, EU *Energy Roadmap 2050*, expert opinion

Table 1: Criticality ratings of shortlisted raw materials

High	High-Medium	Medium	Medium-Low	Low
REE: Dy, Eu, Tb, Y	Graphite	REE: La, Ce, Sm, Gd	Lithium	Nickel
REE: Pr, Nd	Rhenium	Cobalt	Molybdenum	Lead
Gallium	Hafnium	Tantalum	Selenium	Gold
Tellurium	Germanium	Niobium	Silver	Cadmium
	Platinum	Vanadium		Copper
	Indium	Tin		
		Chromium		

A new, preliminary analysis:

What is ‘critical’ for carbon abatement (decarbonization)?

9 technologies*

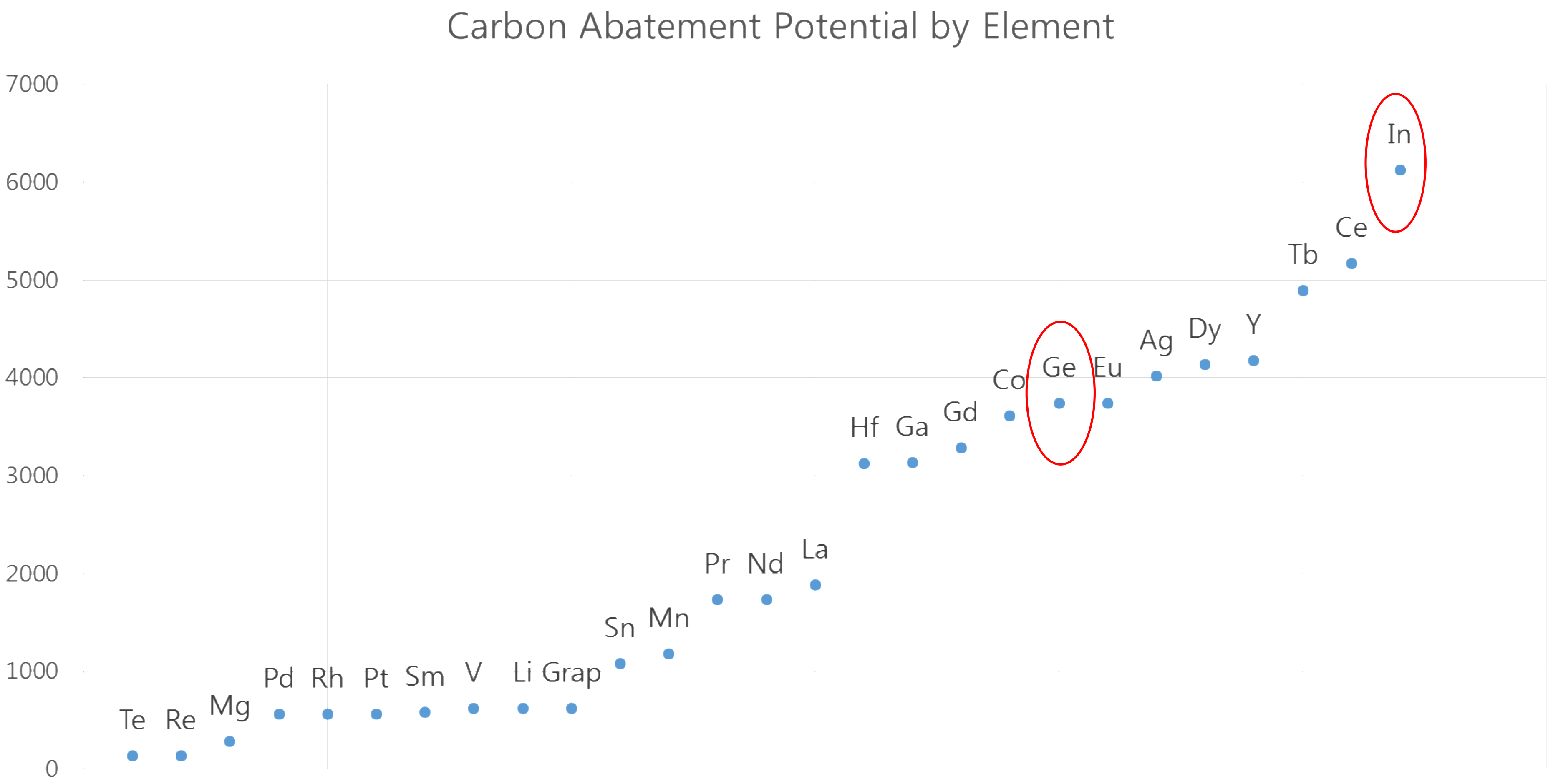
- Photovoltaics
- Wind
- Advanced vehicles, including fuel cell
- Lighting
- Catalytic converters
- Nuclear power
- Gas turbines
- Batteries for electricity storage
- Vehicle lightweighting

*From DOE *Quadrennial Technology Review 2015.*
Did not evaluate hydrogen, electrolysis or thermoelectrics.

Decarbonization

Sector	Technology	Carbon Abatement Potential (MMTCE)	Percentage
Vehicle	Battery EV	38	0.4
	Plug-in hybrid EV	113	1.1
	Hybrid EV	368	3.7
	Fuel cell	8	0.1
Catalytic Converter	Catalytic converter	558	5.6
Vehicle Light Weighting	Vehicle light weighting	287	2.9
Wind	Wind NdFeB	624	6.3
Solar	Solar silicon	724	7.3
	Solar CdTe	135	1.4
	Solar CIGS	129	1.3
Natural Gas	Natural gas turbine	141	1.4
Nuclear	Nuclear generation	2,987	30.0
Electricity Storage	Electricity storage	96	1.0
Lighting	Fluorescent	737	7.4
	LED	3,006	30.2
TOTAL		10,048	100.0

Decarbonization



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27 elements

Rare earths	Other	
Cerium	Cobalt	Platinum
Dysprosium	Gallium	Rhenium
Europium	Germanium	Rhodium
Gadolinium	Indium	Silver
Lanthanum	Graphite	Tellurium
Neodymium	Hafnium	Tin
Praseodymium	Lithium	Vanadium
Samarium	Magnesium	
Terbium	Manganese	
Yttrium	Palladium	

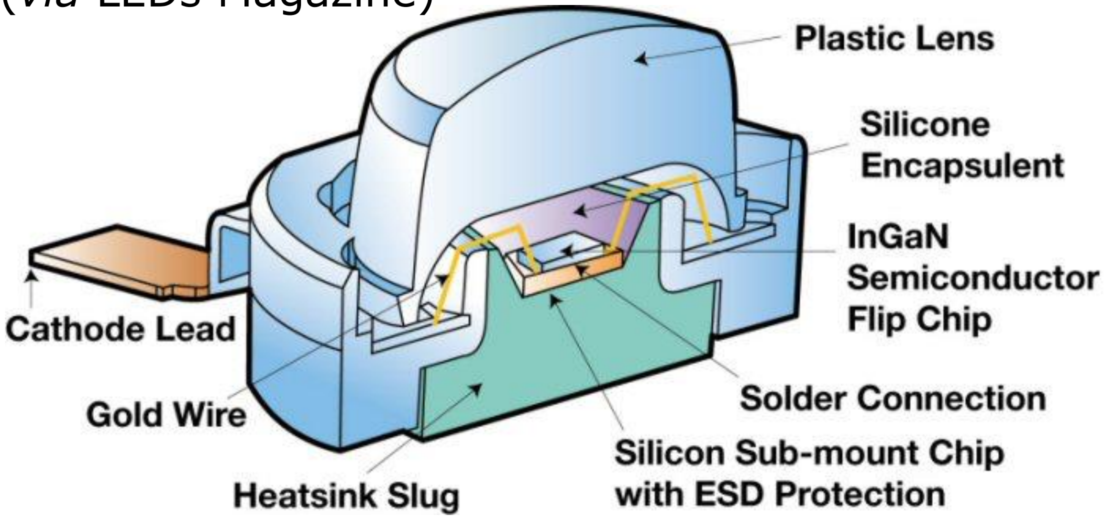
Substitutability: *Summary Results*

Least substitutable →

Material	Score	Material	Score
Ce	3.39	Ag	1.25
Y	3.06	Eu	1.16
Nd	2.26	Gd	1.15
La	2.23	Pd	1.15
Mn	2.23	Rh	1.15
In	2.20	Pr	1.14
Graphite	2.06	Sn	1.05
Dy	1.94	Mg	1.00
Li	1.74	Re	1.00
Co	1.73	Te	0.76
Ga	1.70	V	0.58
Hf	1.50	Sm	0.58
Tb	1.47	Ge	0.41
Pt	1.30		

White LED materials

Lumileds® Luxeon® emitter
(*via* LEDs Magazine)



InGaN LED chip produces blue light

Ceria-doped *YAG* particles embedded in the silicone encapsulant absorb some of the blue and emit a broad spectrum of visible light, producing the desired white light.

First part summary

We anticipate more frequent incidents of criticality over the coming decades because of

- Middle-class population growth
- New technology emergence
- Technological complexity

Criticality is usually temporary

- But it can have long-term impacts.

Better anticipation and faster response are needed

Anacritical materials are an important challenge, too

Produce more

- Diversify and enhance primary production

Waste less

- Use what we do produce more efficiently through improved manufacturing efficiency, recycling and re-use

Use less

- Develop substitutes

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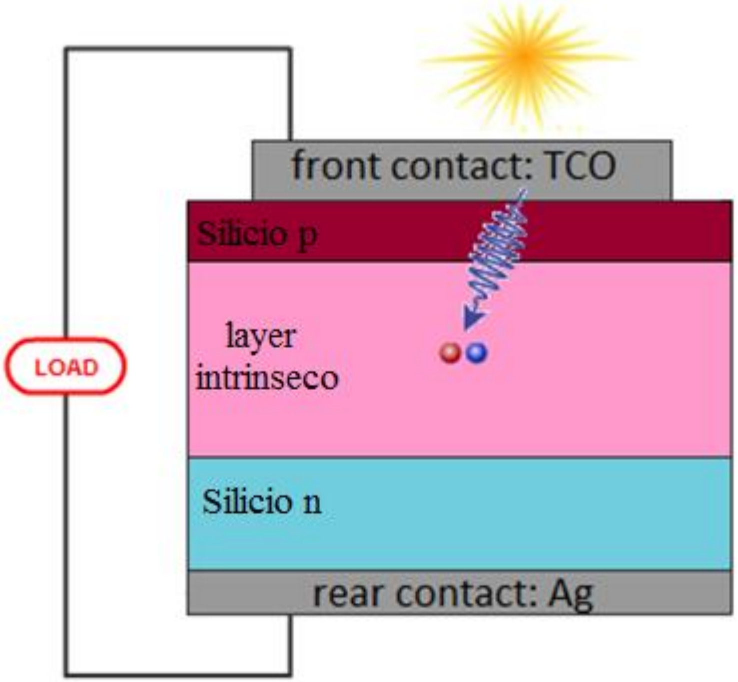
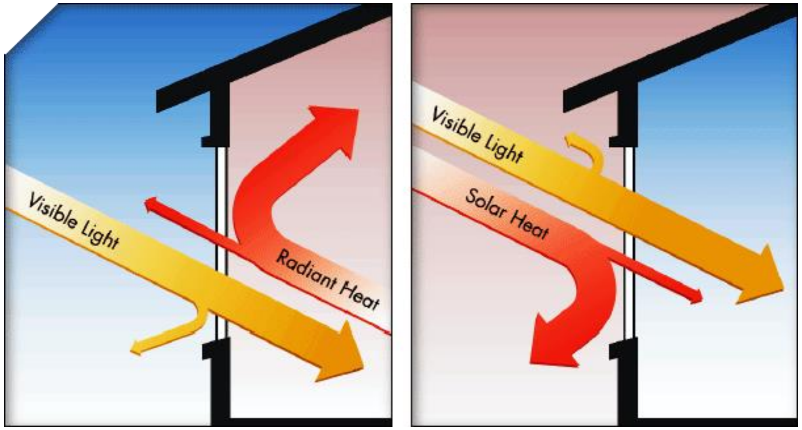
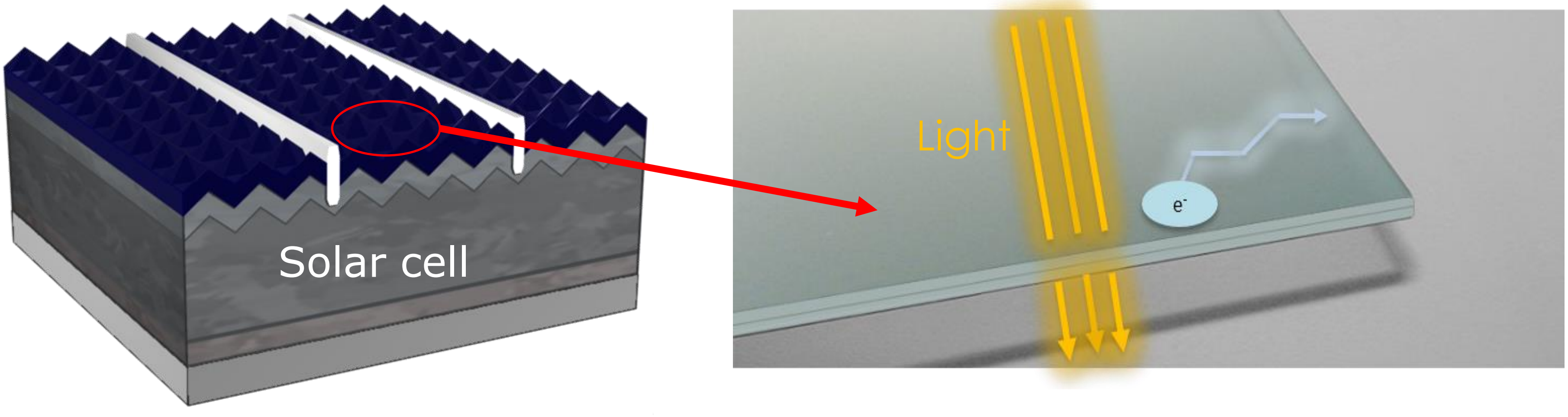
Transparent Conductive Oxides (TCO)



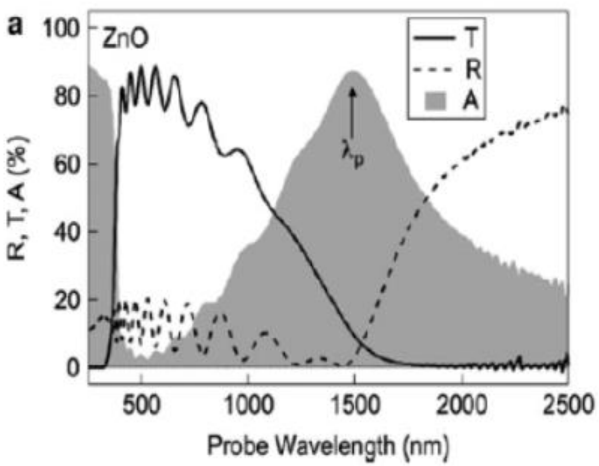


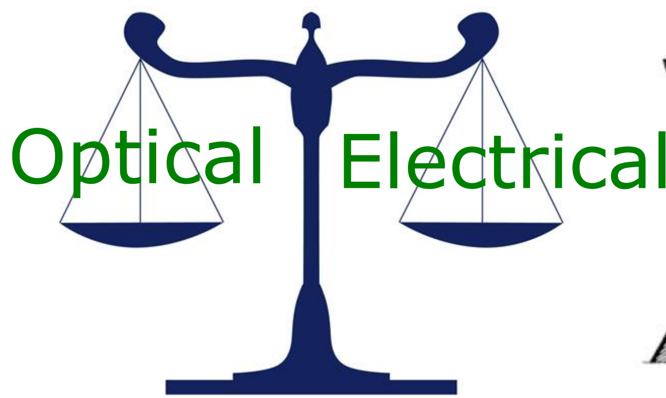
Not only PV: smart windows, touch screens, UV LED, Transparent FET …

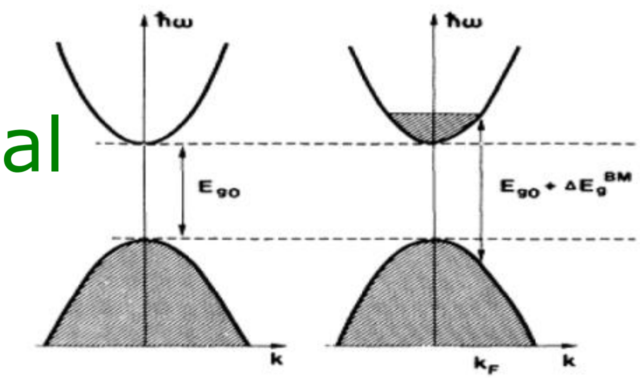
Transparent Conductive Oxides (TCO)



Properties

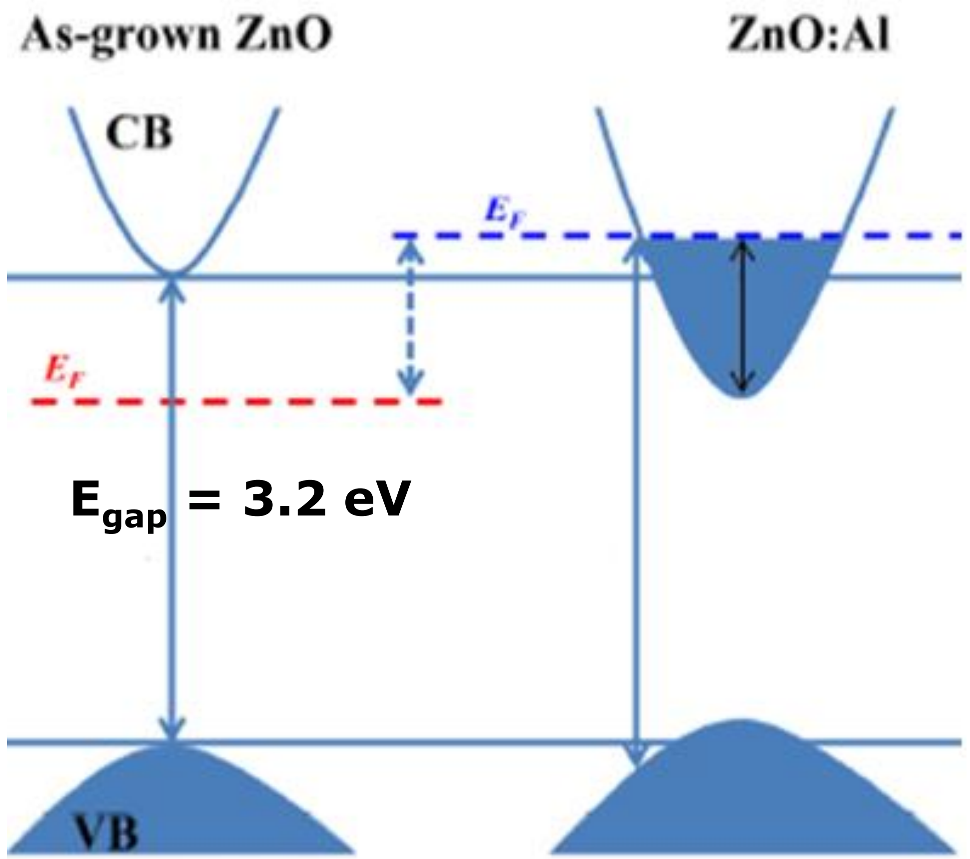






Transparent Conductor Oxides: doping effects

Al^{3+} replace Zn^{2+} so giving an e^- to the CB



E_{gap} = 3.2 eV

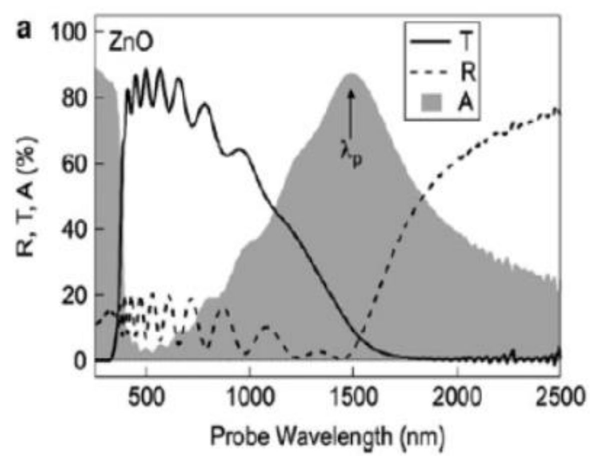
If the elcetron density **n** increases ….

✓ E_{gap} increases (Moss-Burstein effect)

$$\Delta E_{gap\,M-B} = E_{gap} + \frac{h^2}{8m^*}\left(\frac{3}{\pi}\right)^{\frac{2}{3}} n^{\frac{2}{3}}$$

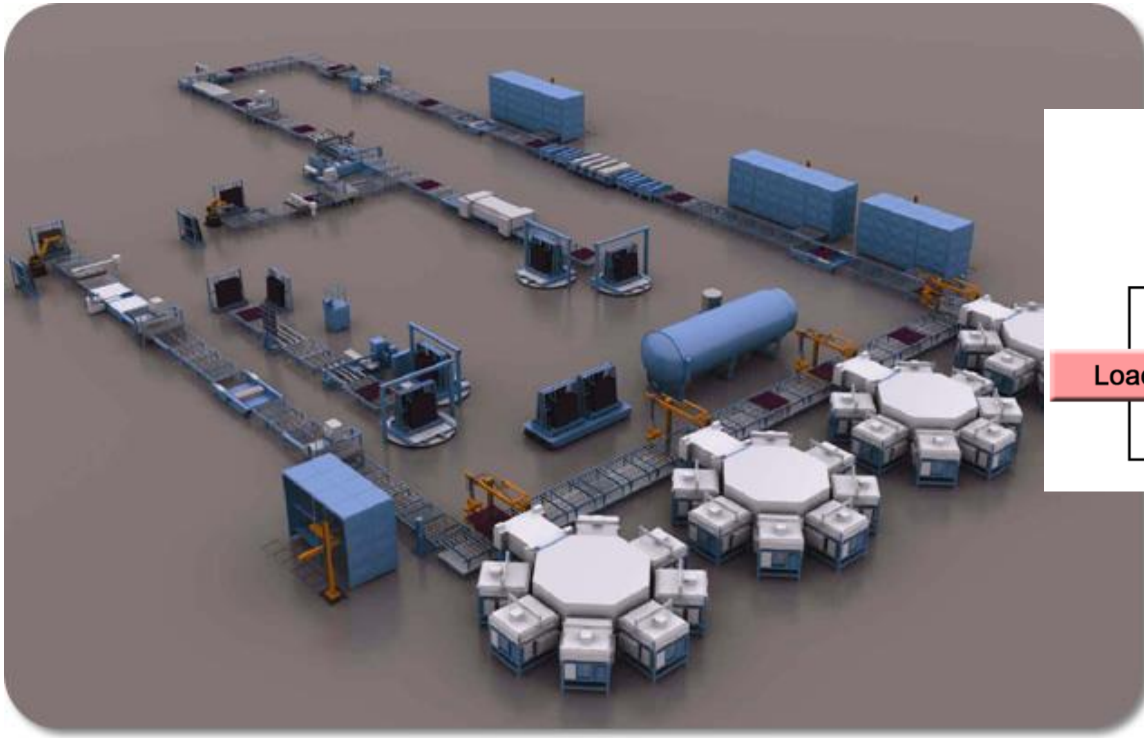
✓Plasma frequency increaes

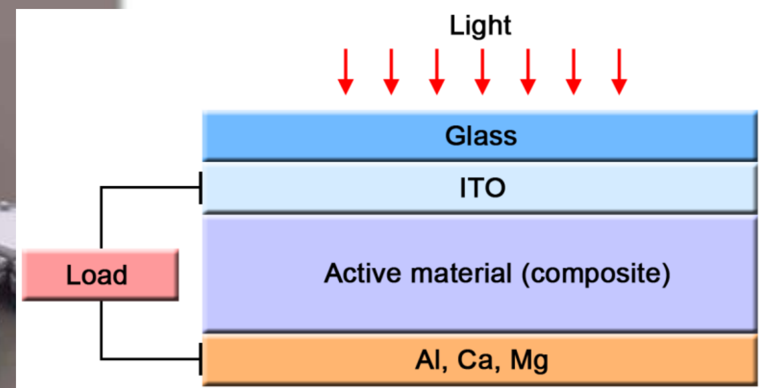
$$\nu_p = \frac{1}{2\pi}\sqrt{\frac{ne^2}{\varepsilon_\infty \varepsilon_0 m^*}}$$

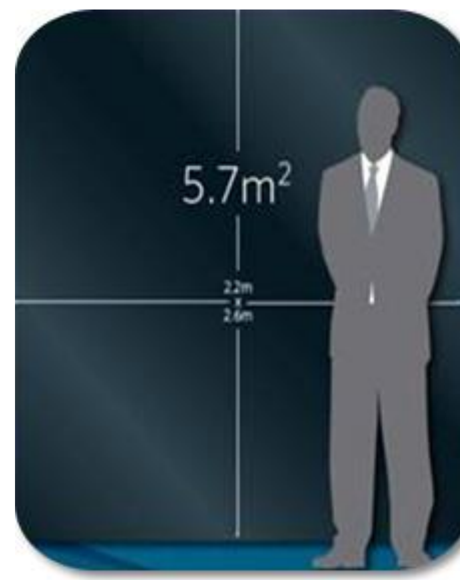


… low electrical resistivity decreases!

TCO for amorphous silicon thin film solar cell







Typical thickness of TCO = 700 nm.

Cost of glass + TCO was 30% of the total cost

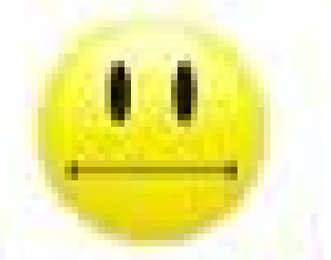
The most used TCO in PV Industry is ITO i.e. In-doped Tin Oxide

ITO properties and drawbacks

In:SnO_2

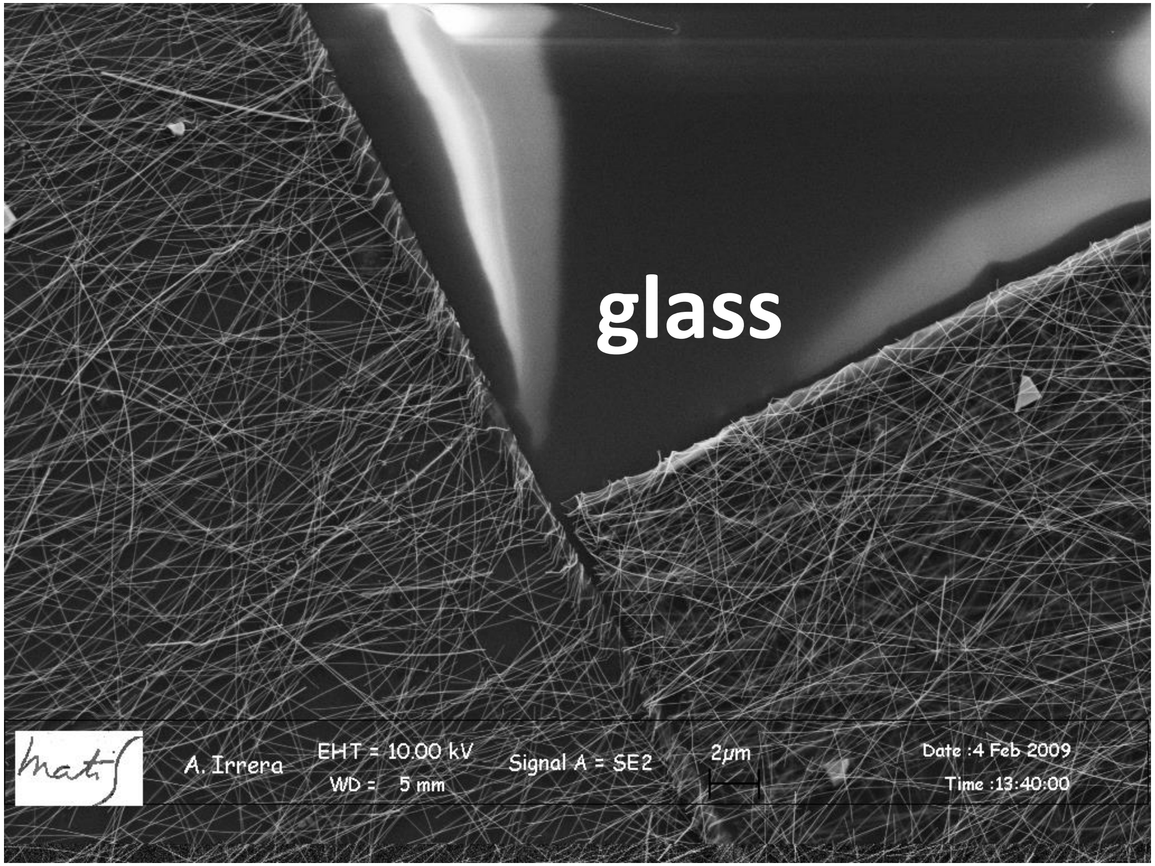
Very good electrical properties

Good optical properties



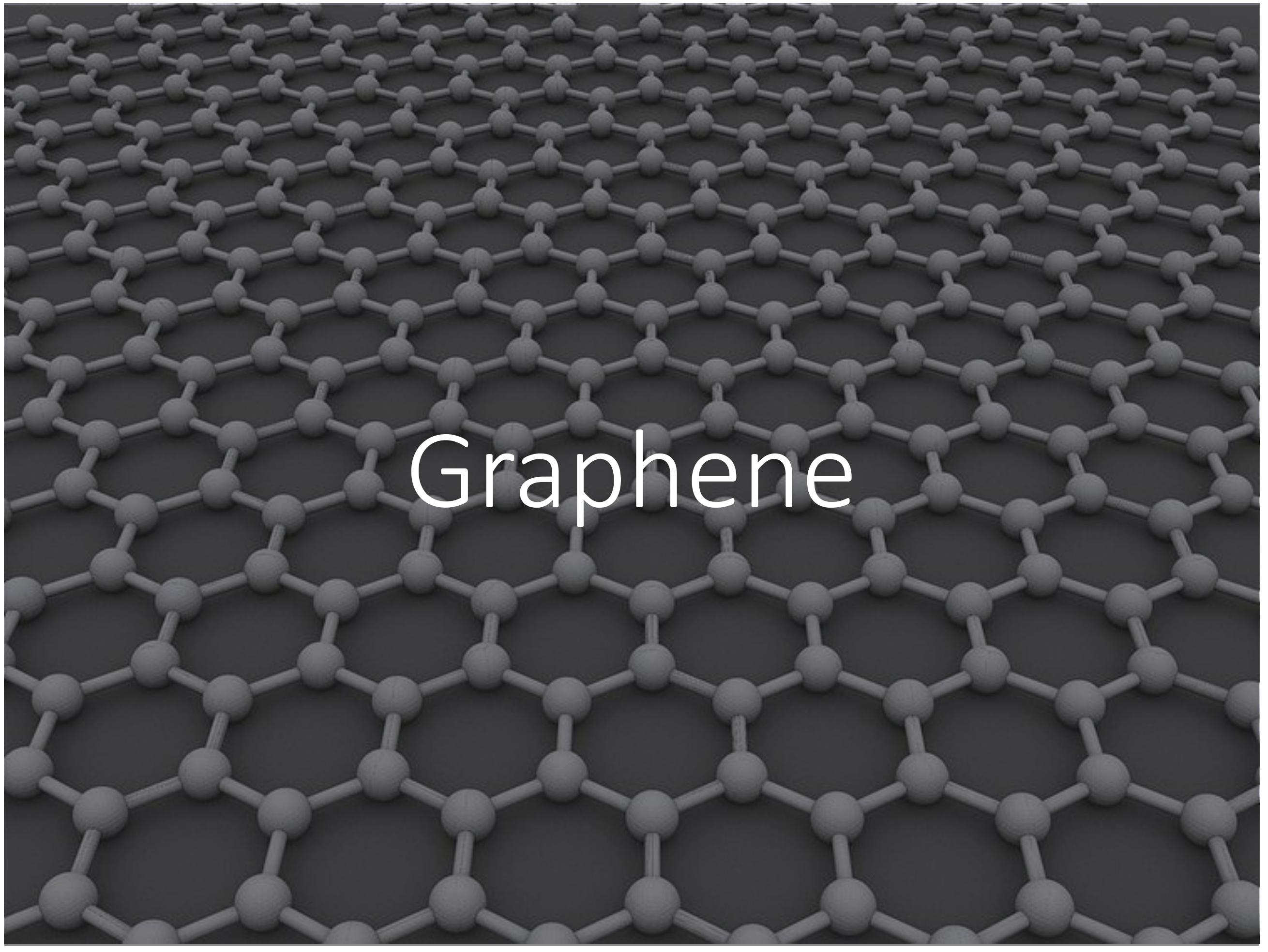
Cost and toxicity

alternative n°1: Silver nanowire network!



drawback: low transparency and very high resistance at the contact points among nanowires

alternative n°2: the "magic" material!



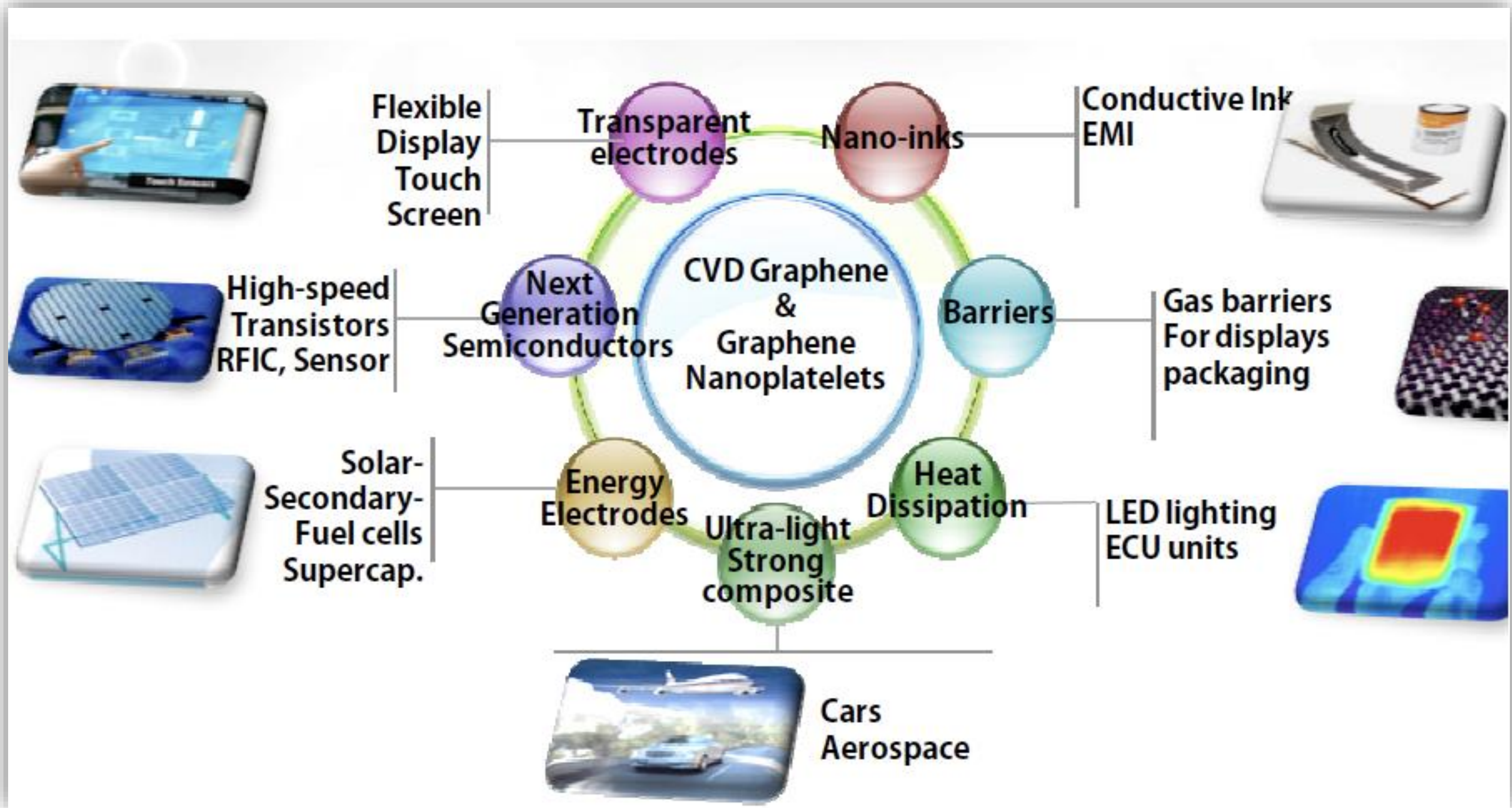
Wonderful structural and electro-optical properties

Physical Properties:		Possible applications
Electronic	Giant "intrinsic" mobility of graphene 2DEG (~2×10^5 $cm^2V^{-1}s^{-1}$) *J.H. Chen, et. al, Nature Nanotechnology 3, 206 (2008).*	•High frequency (GHz -THz) devices • Highly efficient passive components (ultracapacitors). •"Zero energy loss" devices •High conductivity transparent electrode for solar cells
	Large "intrinsic" electron mean free path (~1μm) *X. Du, et al., Nature Nanotechnology 3, 491 (2008).*	
	High specific capacitance (~100 F/g) *M. D. Stoller, et. al, Nano Letters, 8, 3498 (2008).*	
Thermal	Very high thermal conductivity (~50 $Wcm^{-1}K^{-1}$) *A.A. Balandin, et al., Nano Letters 8, 902 (2008).*	
Optical	High transparency (97.7%) in the near infrared and visible range	
Magnetic	Electronic spin-transport detected up to 300 K *N. Tombros, et al., Nature **448**, 571 (2007).*	Spintronics
Mechanical	Young's modulus 0.5 Tpa. *C. Lee, et al., Science **321**, (5887) 385 (2008).*	MEMS, NEMS

Electronic Properties

	Si	Ge	GaAs	4H-SiC	GaN	AlGaN/GaN 2DEG	Graphene
Energy band gap (eV) @ 300K	1.1	0.67	1.43	3.3	3.4	3.4	~ 0
Electron effective mass (m^*/m_e)	1.08	0.55	0.067	0.3	0.19	0.19	~0
Intrinsic Electron mobility ($cm^2V^{-1}s^{-1}$) @300K	1350	3900	4600	800	1300	1500 - 2000	2×10^5
Saturation electron drift velocity v_s (10^7 cm/s)	1	0.6	2	2	3	3	>5
Carrier concentration (cm^{-3})	10^{15}	10^{15}	10^{15}	10^{15}	10^{15}	10^{19}-10^{20}	10^{19}-10^{20}

Graphene, graphene, graphene! What else?



Still very very far from the industrial scalability

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Let's go back to Transparent Conductor Oxides

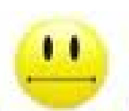
ITO

Indium doped Tin Oxide

$In:SnO_2$

Electrical properties

Optical properties



Cost and toxicity

	AZO (3% Al)	*ITO (10 % In_2O_3)*
T_{fusion} (°C)	2000	1400
Gap (eV)	3.36	3.78
ρ (10^{-4} Ωcm)	**980**	**1.28**
T(% at 550 nm)	≥ 85%	≥ 80%

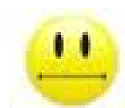
Indium free TCO

AZO

Aluminum doped Zinc Oxide

Al:ZnO

Electrical properties

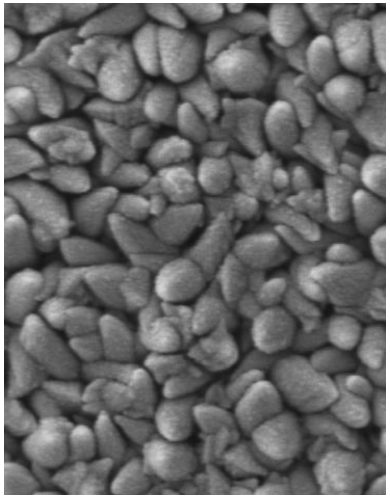


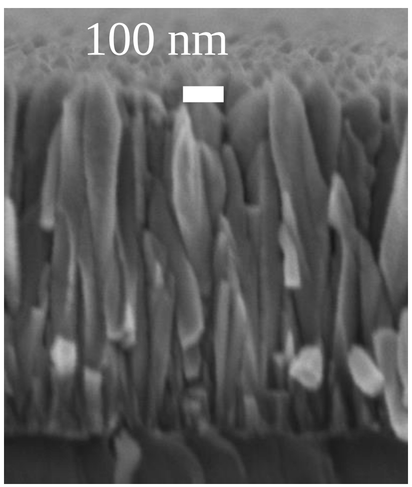
Optical properties

Cost and toxicity

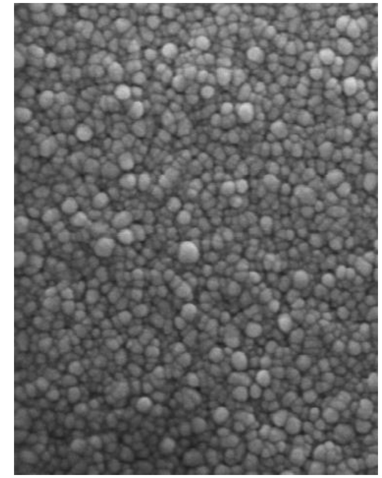
Tony Terrasi: "Energy Materials", EPS-SIF Joint School on Energy, Varenna (Italy), July 21-26 2017

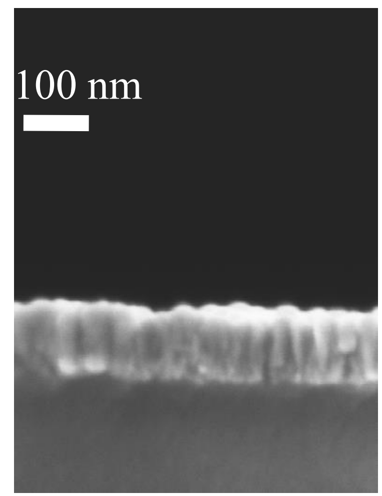
Transparent Conductive Oxides (TCO)





We need to eliminate Indium and to reduce the thickness of the TCO layer from 700 nm to 100 nm or less, by keeping good electrical conductivity





SEM plan views and X-sections of TCO films

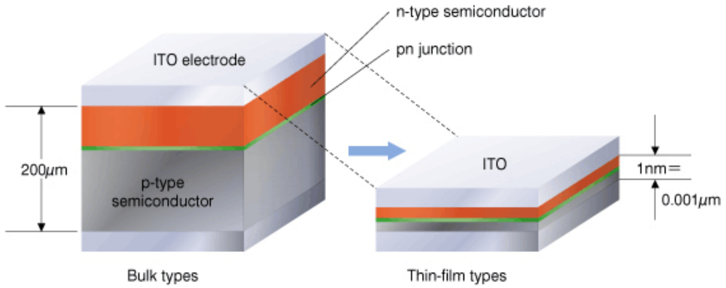
How to reach new requirements?

- Low cost, non-toxicity

e.g. → **Indium free materials,**
polymer-metal hybrid structures, metal nanowires, graphene.

- Flexibility, lightness and cost reduction

e.g. → **Thickness reduction (d)**



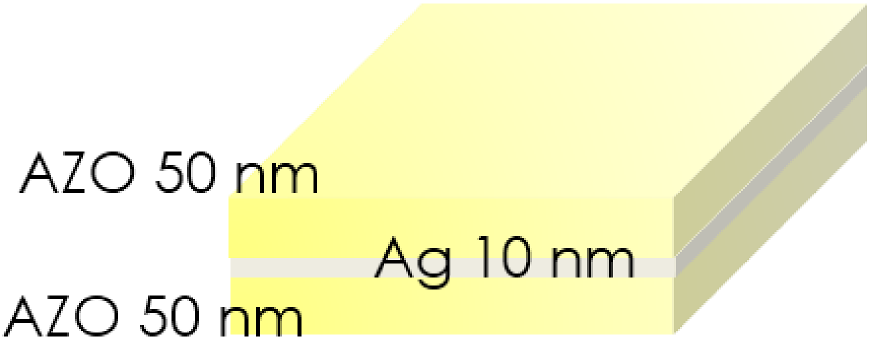
contraindication

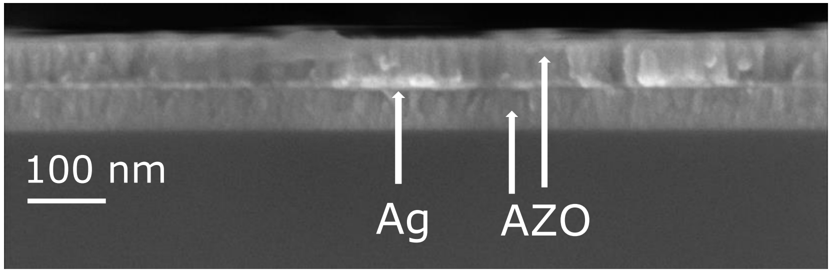
$$R_s = \frac{r}{d} \longrightarrow$$ Lost of good electrical properties

New Materials: AZO/Ag/AZO multilayers

Advantages

- *In free*
- *Thin structures*
- *Industrial compatible growth: sputter deposition*





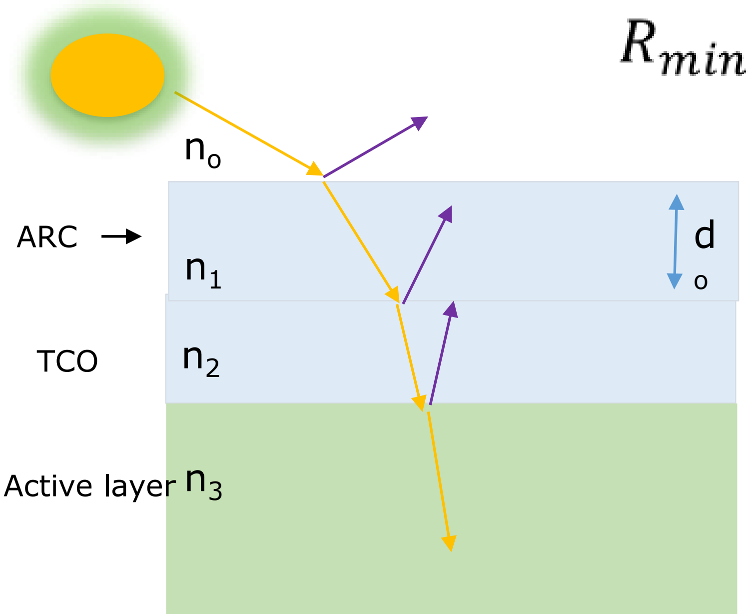
- *Good transparency and metallic conductivity*

	ρ (10^{-4} Ωcm)	T (%)
ITO (700nm)	1.28	80
AZO (700nm)	980	82
AZO (100 nm)	1270	84
AZO/Ag/AZO (100 nm)	0.7	79

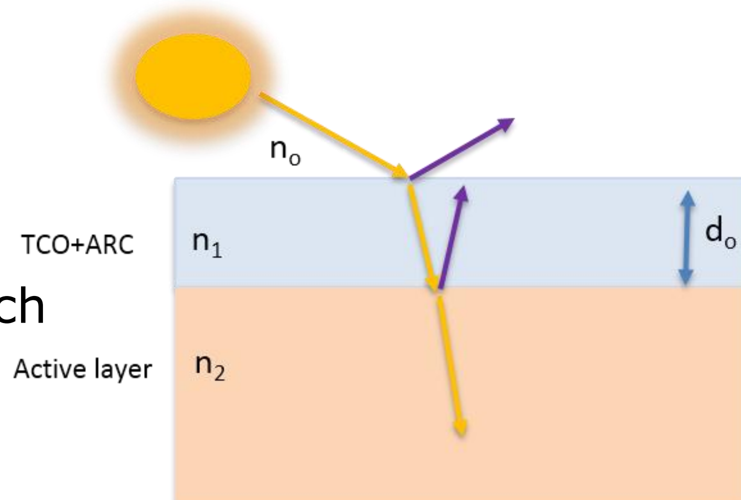
Not only electrical requirements: anti-reflection films

Best choice

$$R_{min} = \frac{(n_1^2 - n_o n_2)^2}{(n_1^2 + n_o n_2)^2} \longrightarrow d_o = \frac{\lambda}{4n_1}$$

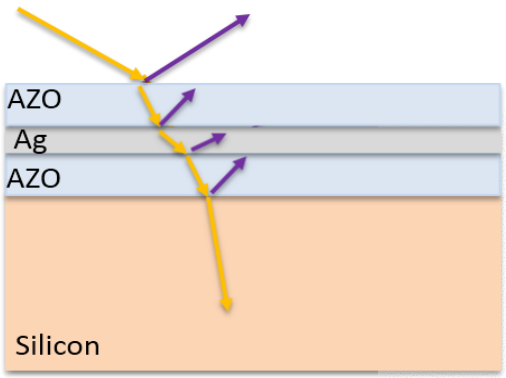


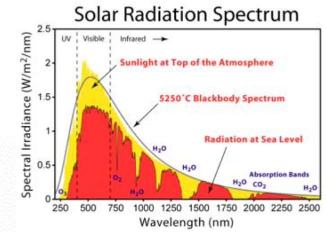
New approach

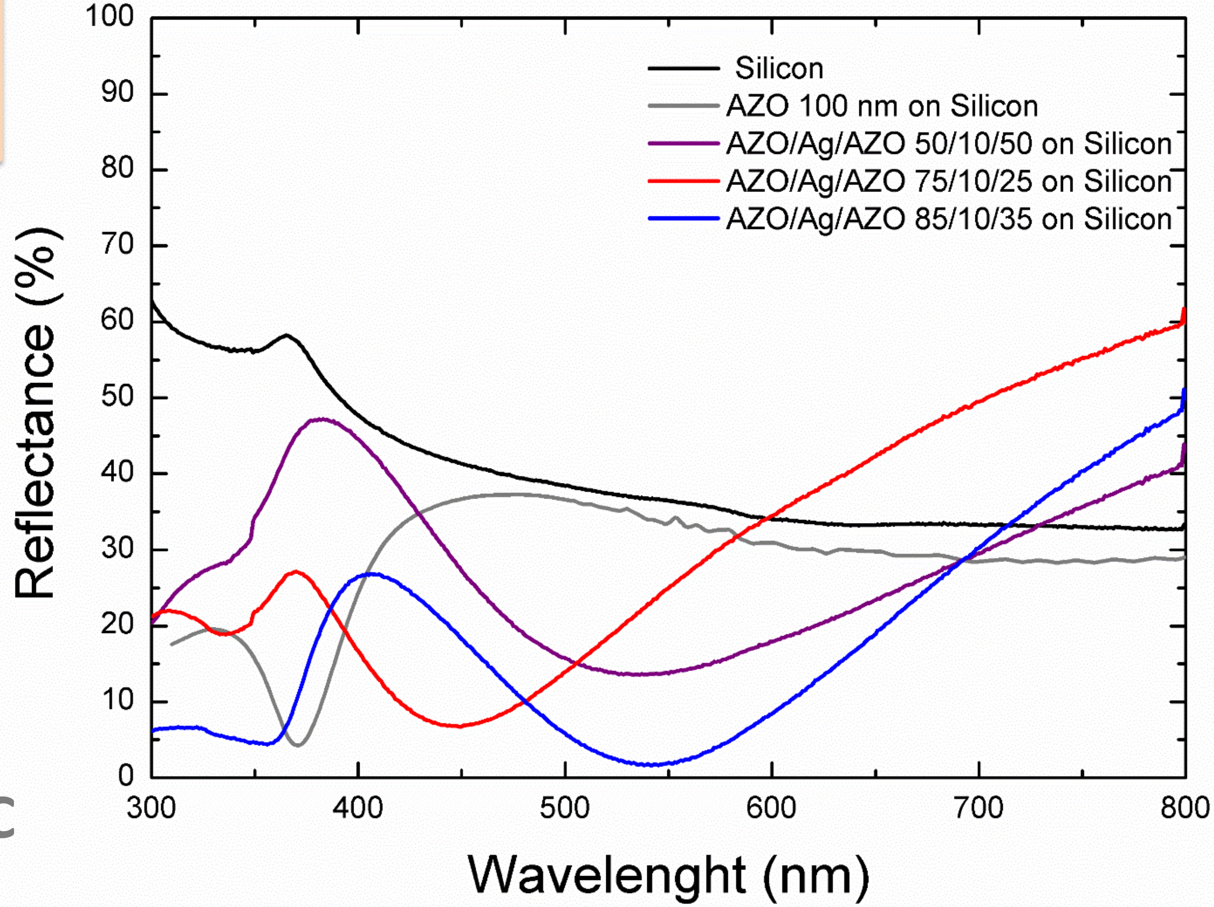


also architectural needs

Anti-reflecting coating: AZO thickness effect



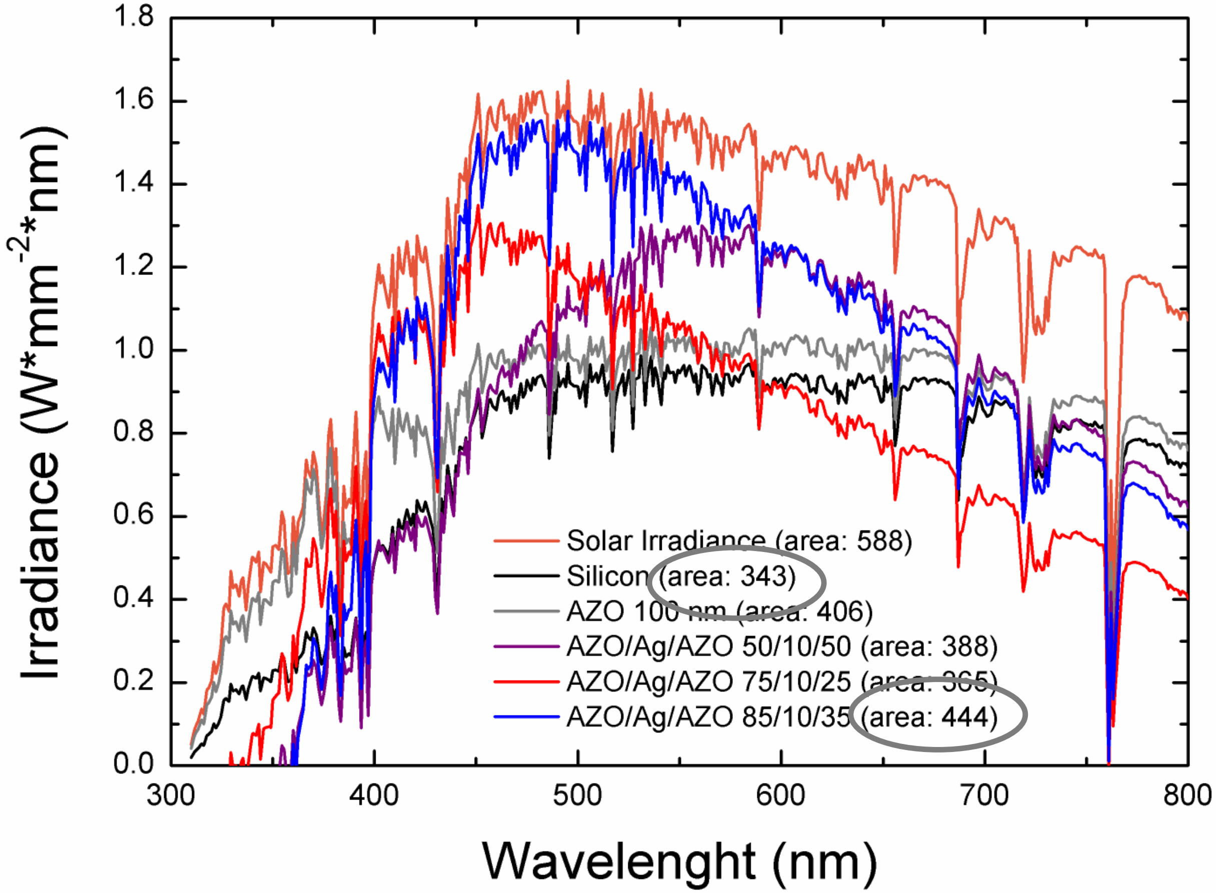


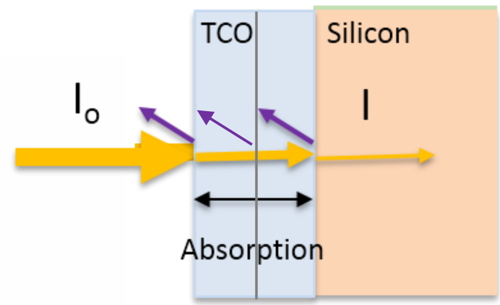


TCO acting as ARC

Effect on a solar cell

$$I = I_o$$





Light reaching the Si substrate increases of 17.5 %

Main conclusion at this stage of research is that AZO/Ag/AZO could replace standard TCO + ARC material and technology

Outline

1. Criticality in Materials

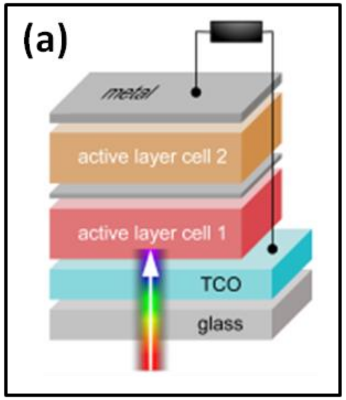
2. Criticality in Energy Materials

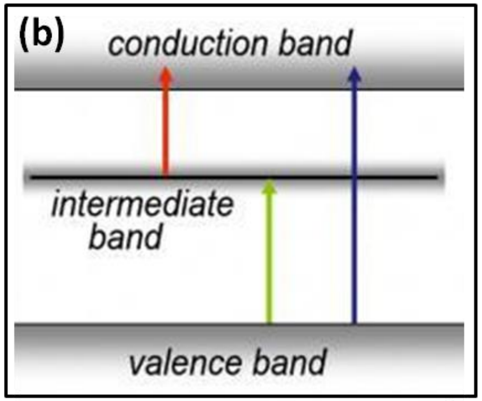
Two examples of research in
energy-related material science

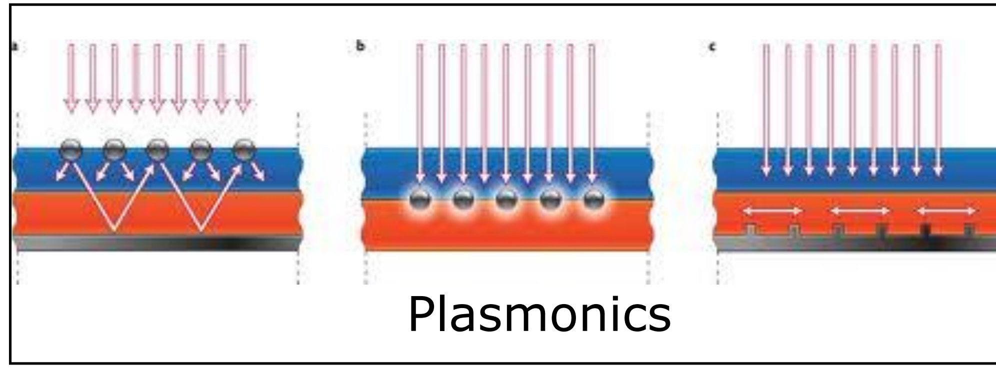
3. How to replace a material:
   Transparent Conductor Oxides

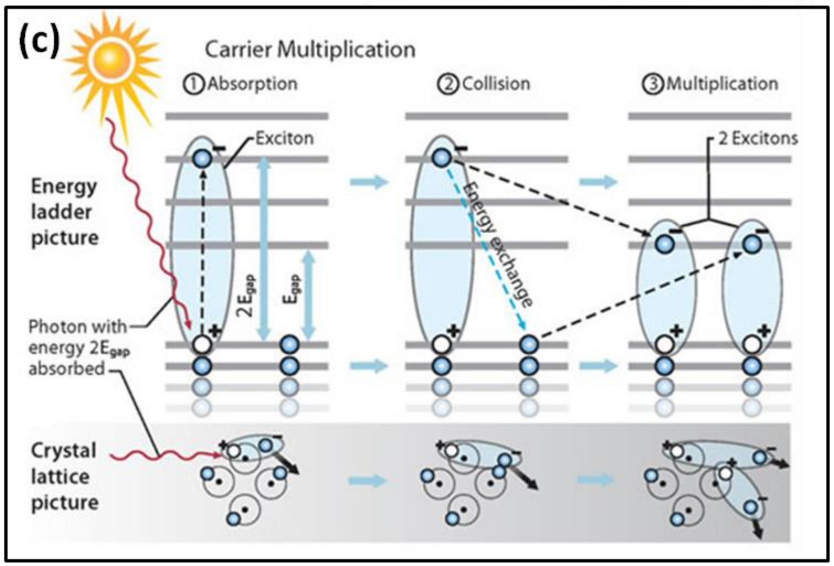
4. How to modify material properties:
   Quantum confinement effects in Ge Nanostructures

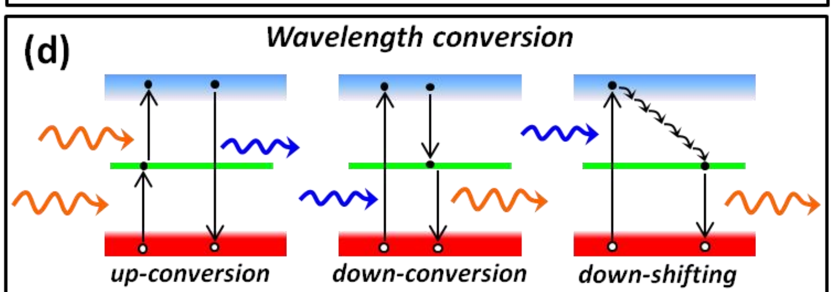
Nanostructures for Efficient Light Harvesting

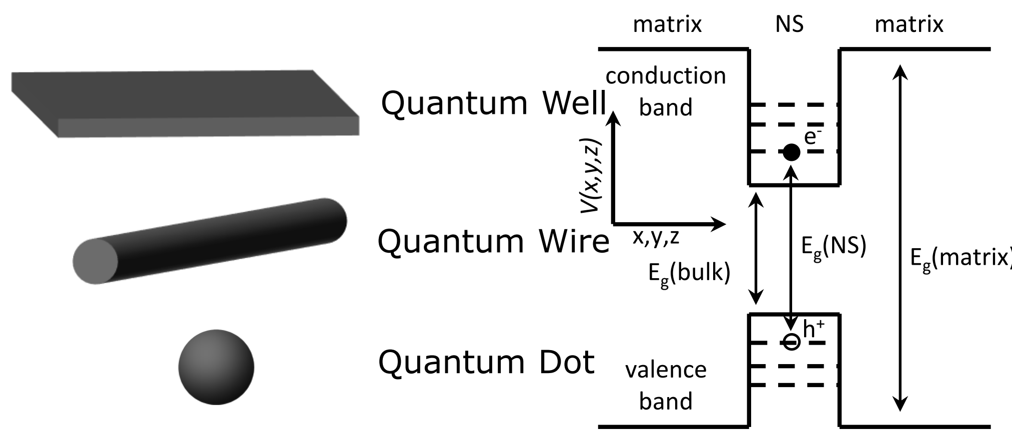










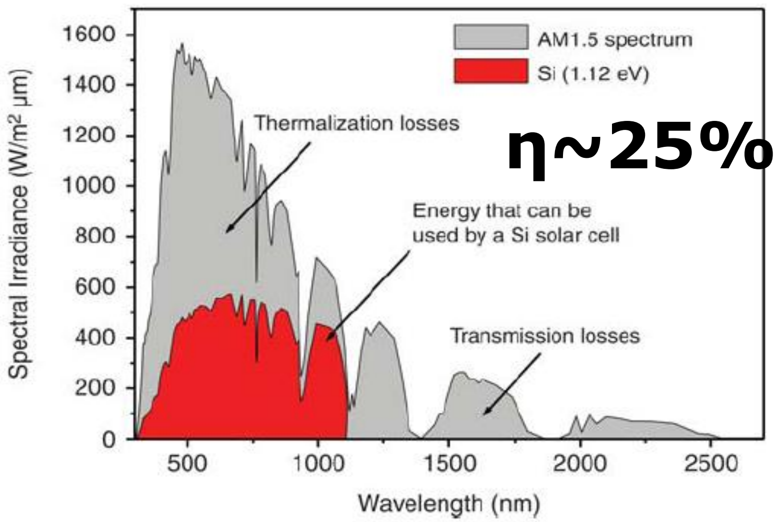


$$E_g(NS) = E_g(bulk) + \frac{\pi^2 \hbar^2}{2m^* L^2}$$

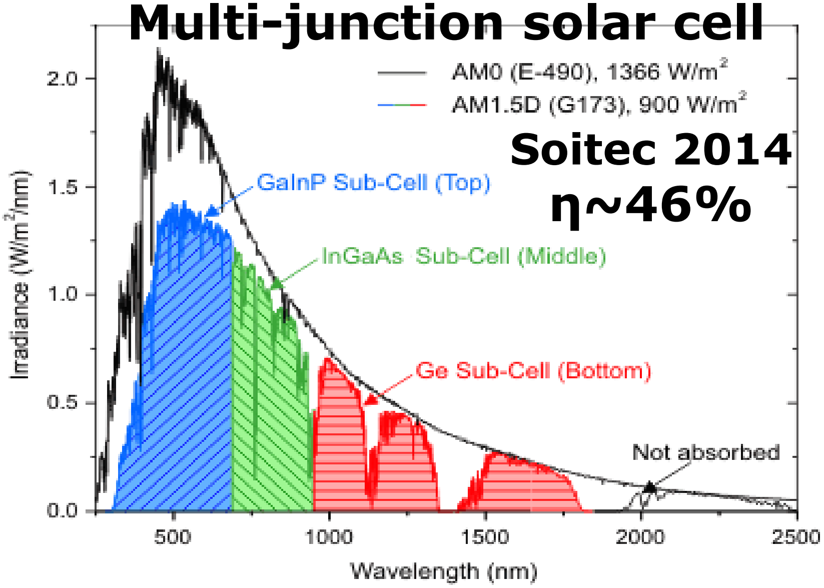
Bandgap tuning in nanostructures

Quantum confinement effect

Single-junction solar cell



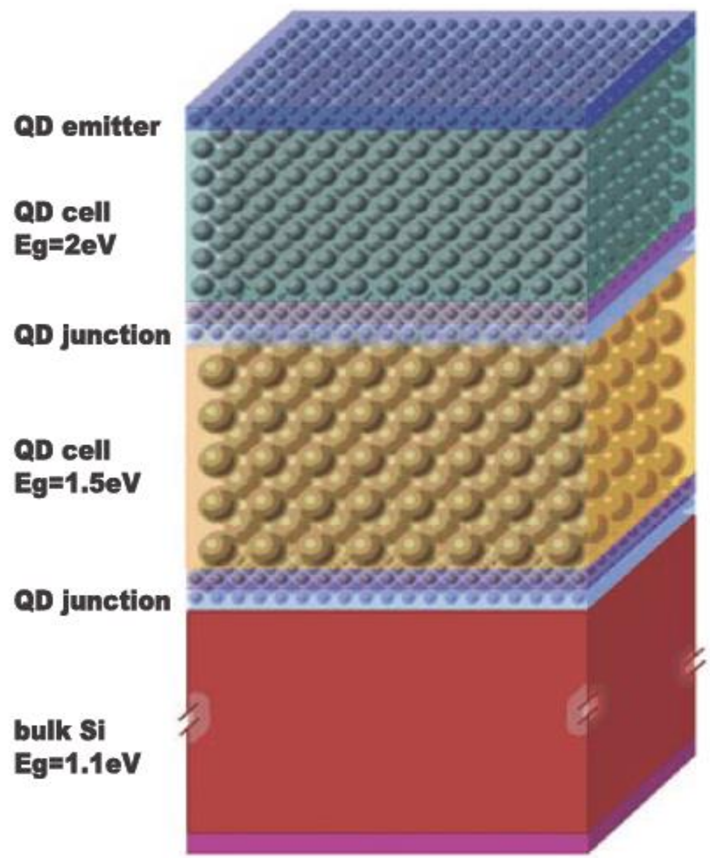
Multi-junction solar cell



F.Dinroth, S.Kurtz, **MRS bulletin, 32, 230 (2007)**

Optical bandgap tunable in QD → filling all the solar spectrum!

All Si tandem solar cell
UNSW group Green et al., 2005



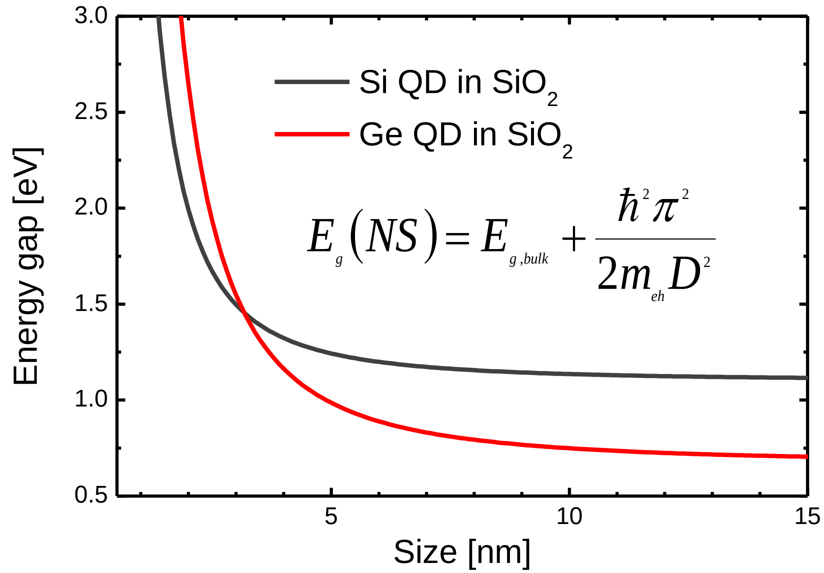
Are Ge nanostructures better than Si ones?

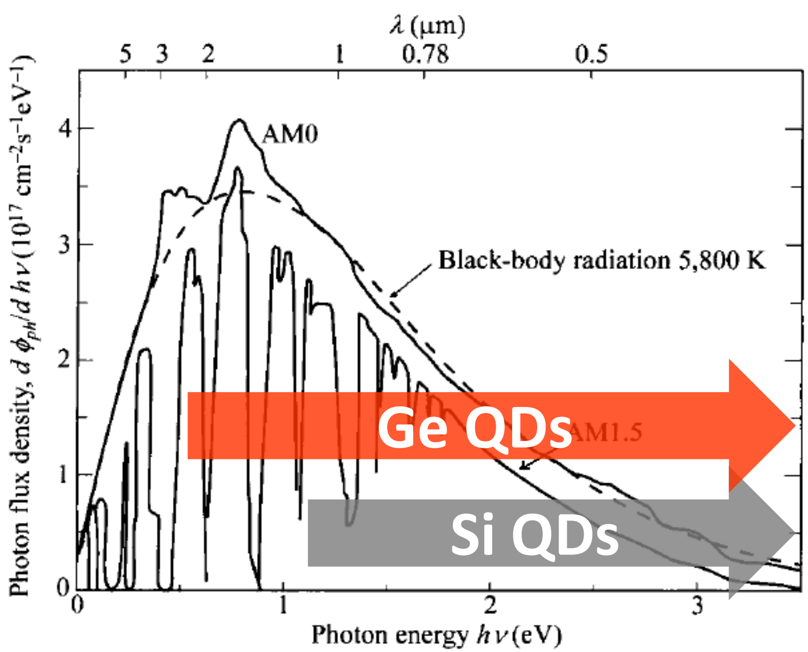
Large Bohr exciton radius

Semiconductor	Band gap size (eV)	Dielectric constant	Exciton Bohr radius[a] (Å)
Si	1.12	11.4	~49
Ge	0.66	15.4	~177

Cullis et al. - *J. Appl. Phys.*, **82**, 909 (1997)

Park et al., Phys. Rev. Lett. **86**, 1355 (2001)



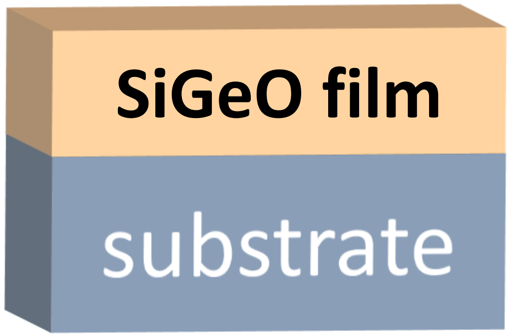


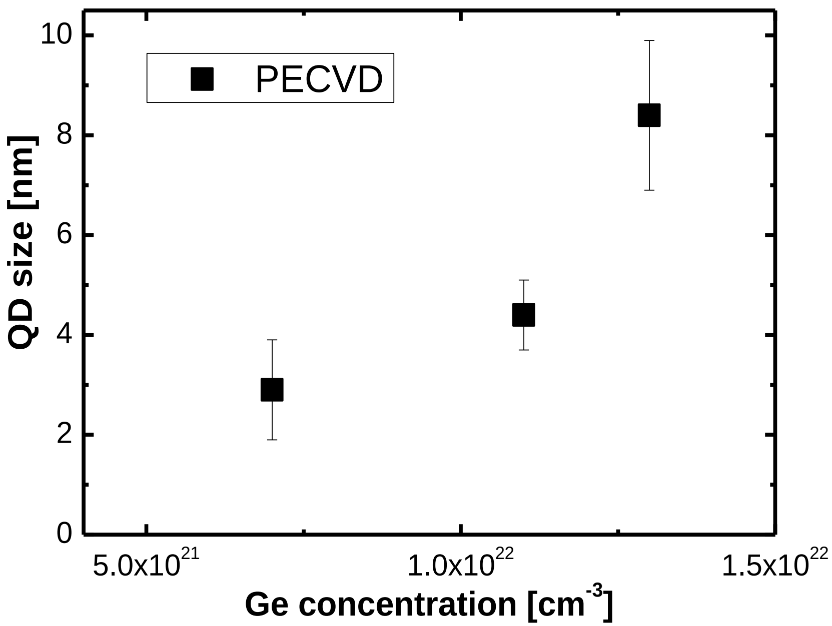
Light Absorption investigation in Ge NS in "confinement" regime

How to prepare Ge nanostructures

PECVD

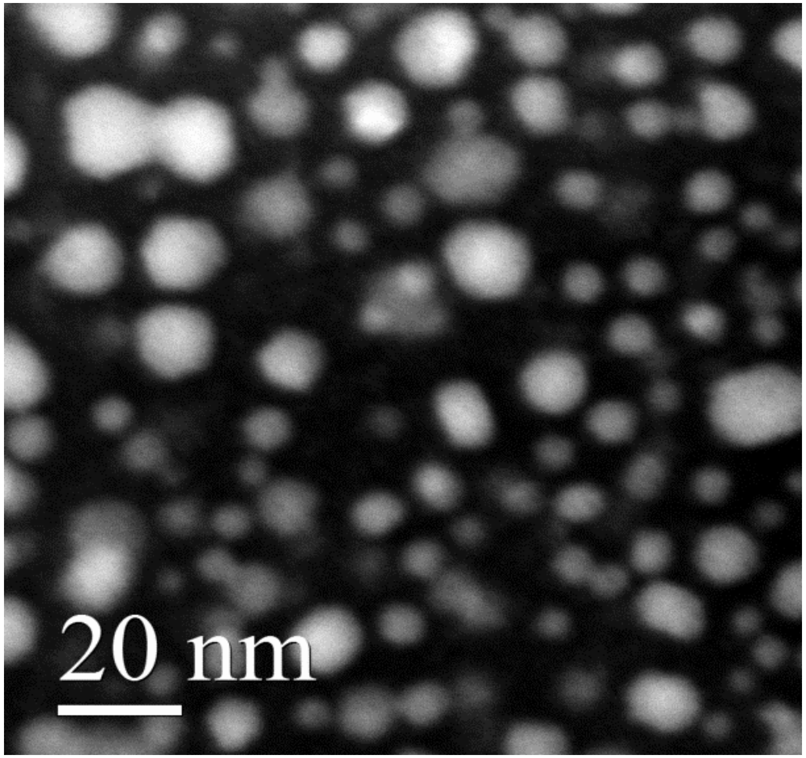
(deposition 250°C: 8 - 20% Ge)
(600-800°C annealing in N_2)





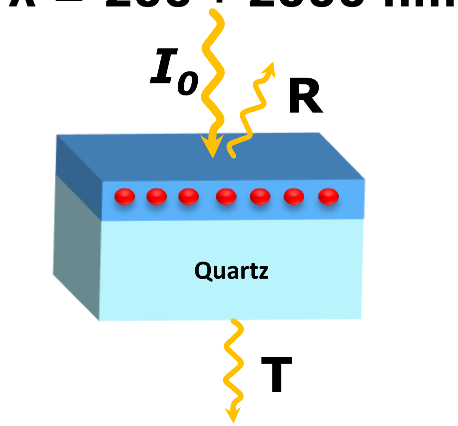
TEM analysis

Ensemble of Ge QDs

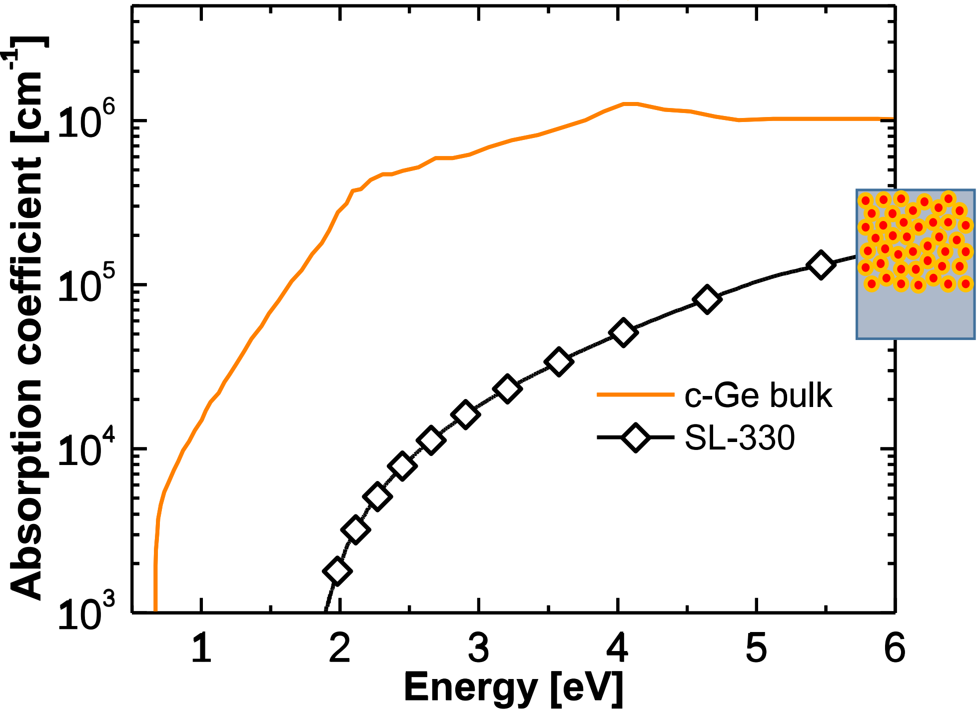


Broad size distribution

Optical absorption Coefficient

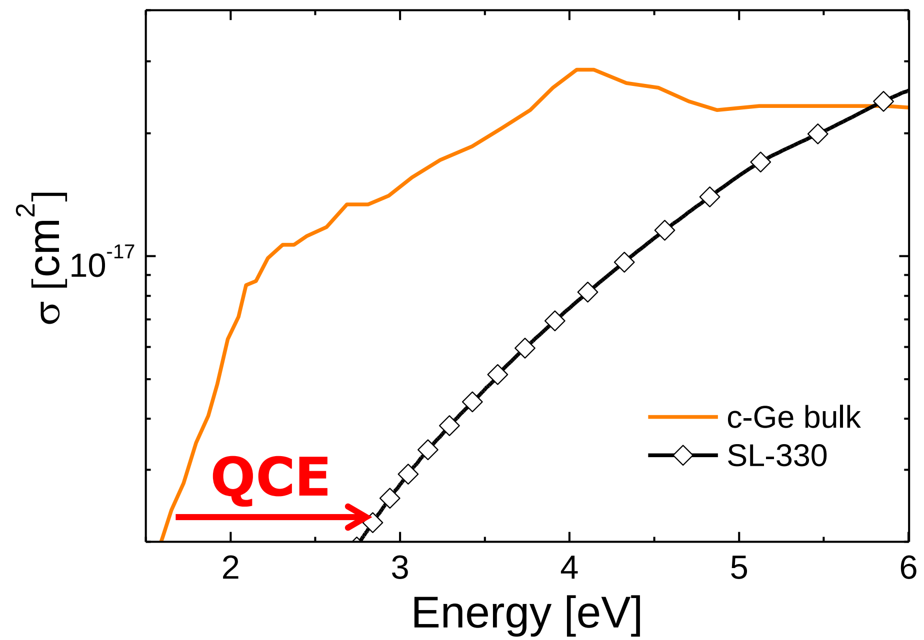


$$\alpha = \frac{1}{d_f} \ln \frac{T^{Quartz}(1 - R^s)}{T^s}$$



Absorption cross section (σ): photon absorption probability per Ge dose

$$\sigma = \alpha \frac{d_f}{D_{Ge}}$$

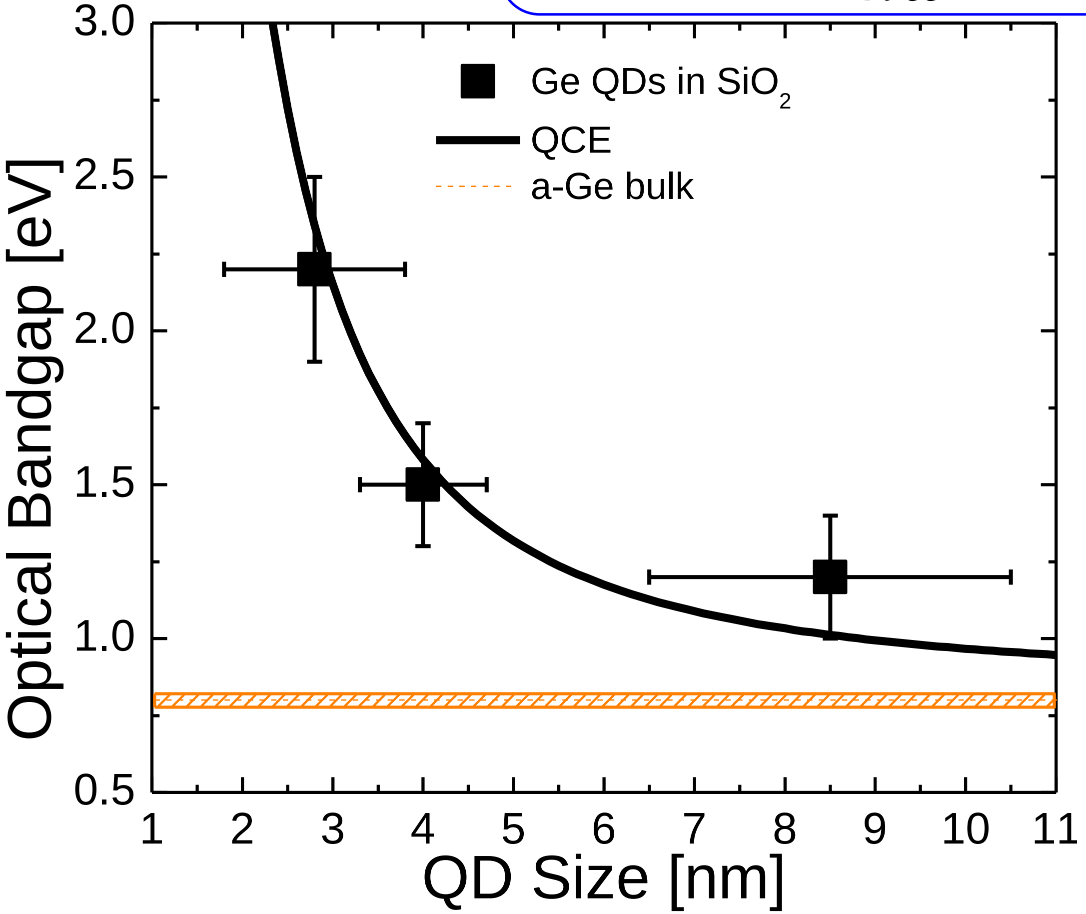


QCE: ensemble of Ge QDs

Extraction of optical properties (E_g and B_{Tauc})

Tauc Formula:

$$\alpha(\omega) = \frac{B_{Tauc}}{\hbar\omega} \cdot \left(\hbar\omega - E_g^{opt}\right)^2$$

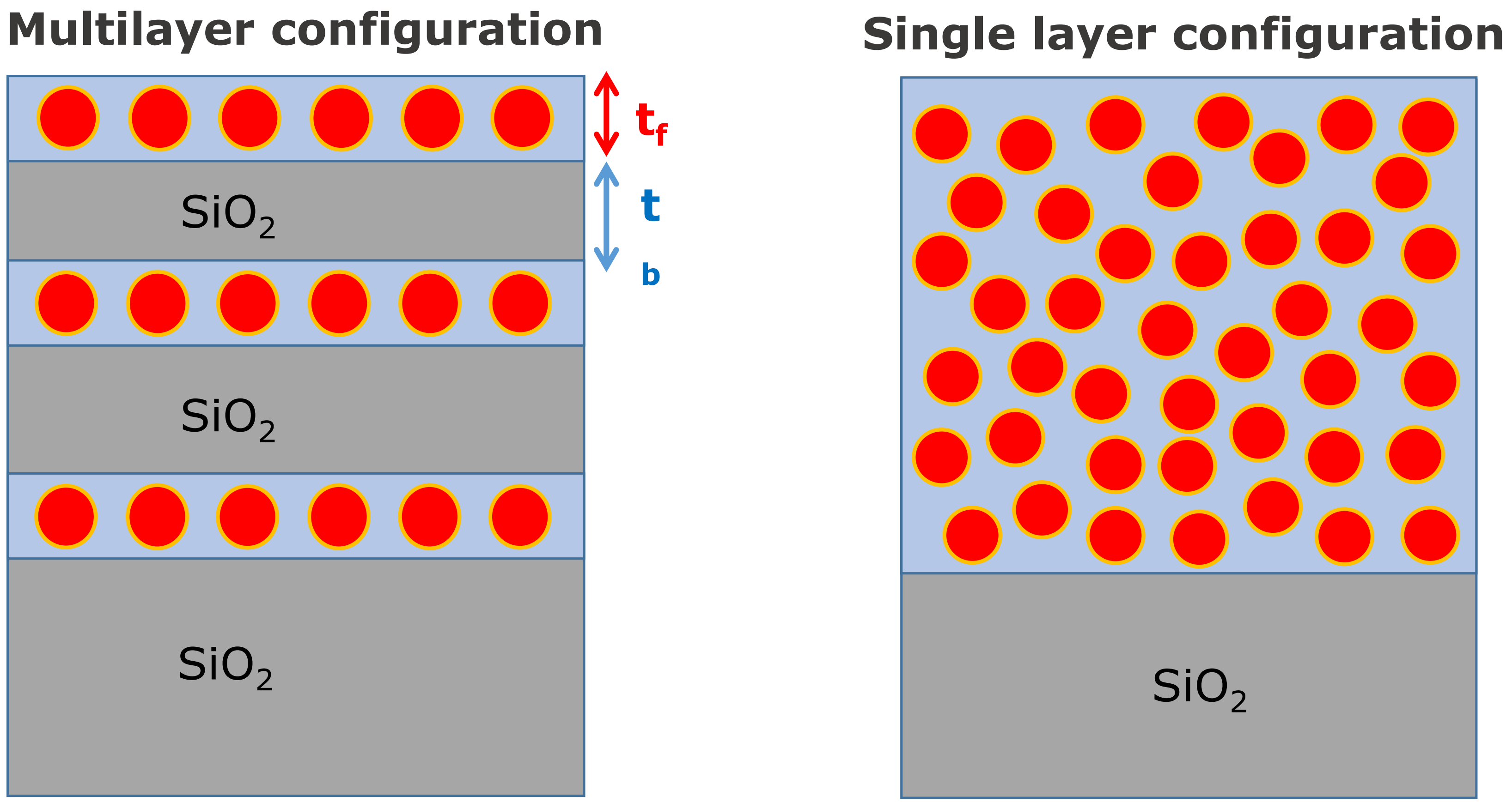


Optical bandgap of Ge QDs shows a clear dependence with QD size in agreement with QCE

PAY ATTENTION TO B_{TAUC} ! It gives you the light absorption efficiency of your material

Controlling your material: Multilayer approach

Multilayer approach for narrowed size distribution

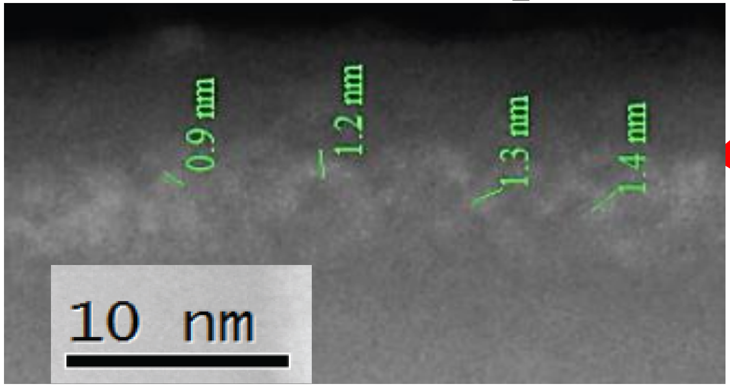


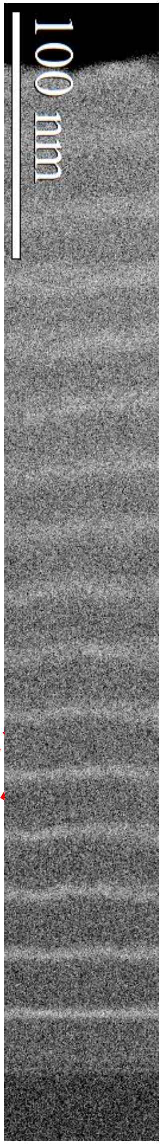
Ge quantum dots in Multilayer configuration

TEM analysis

PECVD

(deposition 250°C: ~10% Ge per layer)
(thermal annealing: 800°C-1h- in N_2)

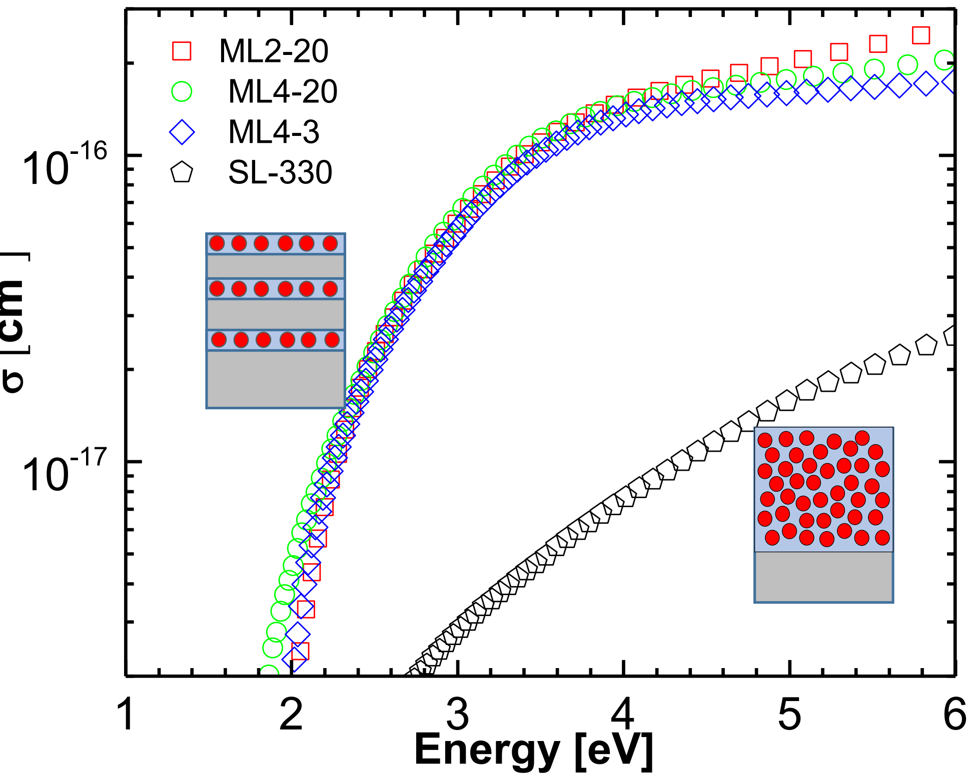


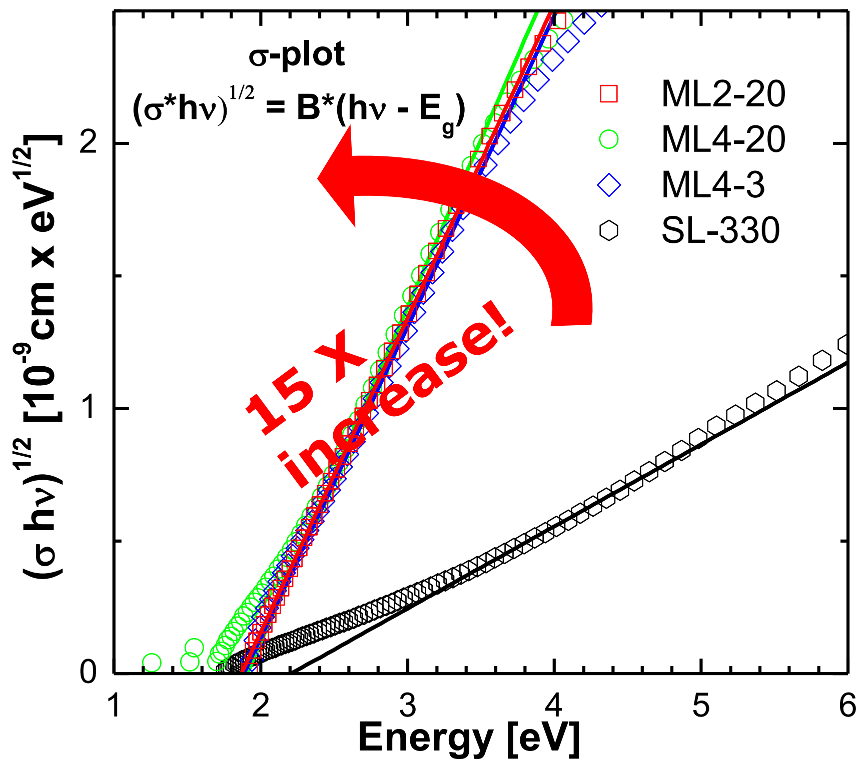


Sample	t_f [nm]	t_b [nm]	N	$2r$ [nm]
ML2-20	2	20	15	2
ML4-20	4.5	20	4	1.7

- **Ge QDs already present in as deposited samples**
- **Narrow size distribution and very small Ge QDs (~2 nm)**
- **QD size does not depend on SiGeO thickness**
- **After annealing QDs remain amorphous, due to their small size**

Light absorption: Single layer vs Multilayer





$$slope \sim B_{Tauc} \sim \frac{2m\omega_i}{e^2\hbar} |< \psi_i \mid e_z \mid \psi_0 >|^2 = O_{strength}$$

The slope is directly proportional to B_{tauc} which, on turn, is proportional to the absorption of the light.

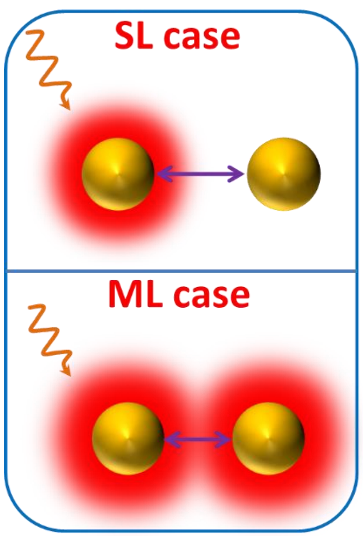
Why this Enhancement of the absorption?

JLDOS (Joint Localized Density of States) between quantum dots with shorter interparticle spacing increases!

M.Logar et al, Nano Lett. 15, 1855 (2015)

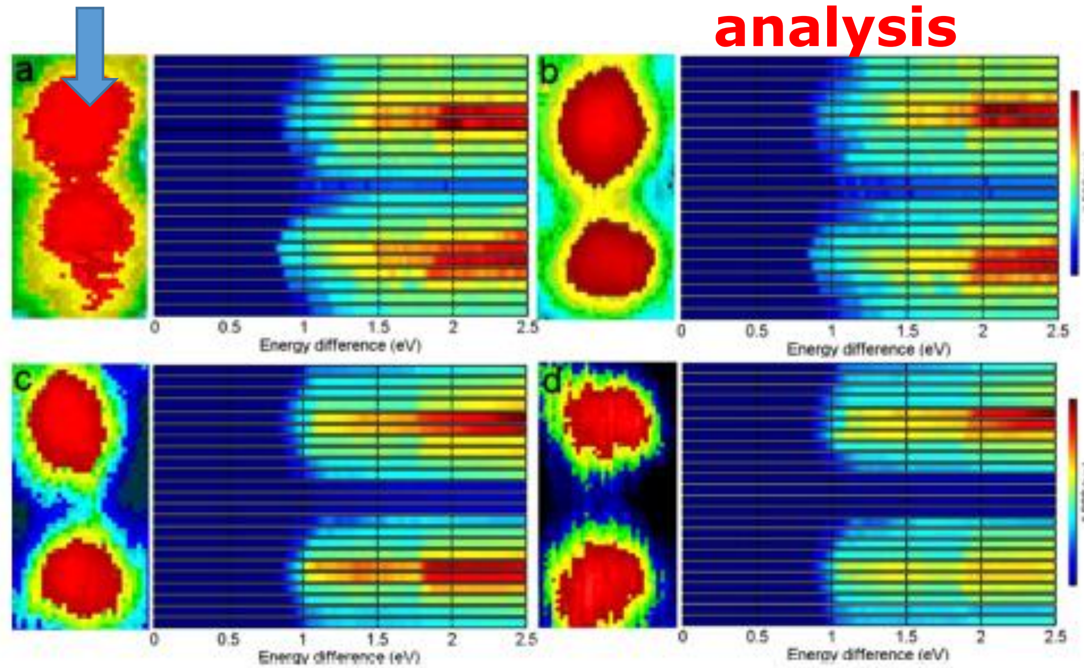
Sample	*S_2S distance [nm]*
Multilayer	**1.2±0.4**
Single layer	**2.9 ±1**

Shorter distance QD-QD in ML



PbSe QDs (~1-2 nm)

EELS analysis



$$\alpha(\omega)=\frac{4\pi^2e^2}{ncm^2\omega}J_{cv}(k)\cdot|M_{cv}|^2$$

JLDOS

To increase JLDOS is equivalent to increase the amount of the absorbing material !!!

Second Part Summary

1. R&D in Material Science is one of the key aspect to face in a successfully way future criticalities in the field of energy production, storage and smart use.

2. Look at today available materials as well as to feasible and reliable technologies to have fast and industrially scalable improvements.

3. Do not neglect future and fancy materials (e.g. graphene) and technologies (e.g. quantum technologies) to solve incoming criticalities.

Thanks for your attention

&

Thanks to:

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2) Roderick Eggert (Colorado School of Mines, USA)

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(Ph.D. student)

TCO

Rosario Raciti
(Post Doc)

Ge QD