Breaking Efficiency and Stability Barriers of Perovskite Solar Cells via Lattice Battery Solar Cell

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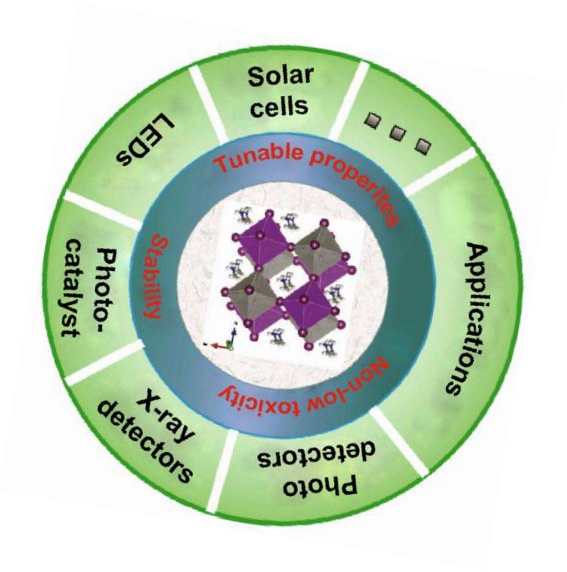
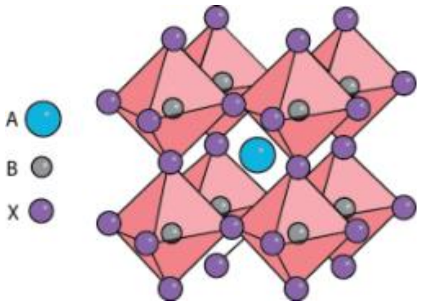
Outline

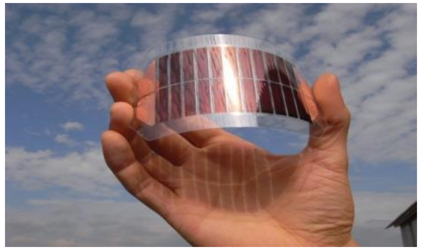
1. Superior properties of metal halide perovskites
2. Anomalous phenomena: conflict to semiconductor theory
3. Theory of lattice energy reservoir
4. Physical effects of lattice energy reservoir
5. Lattice battery solar cell
6. Summary

1. Superior properties of metal halide perovskites

MHPs have unique soft lattice and mixed ionic-electronic conduction





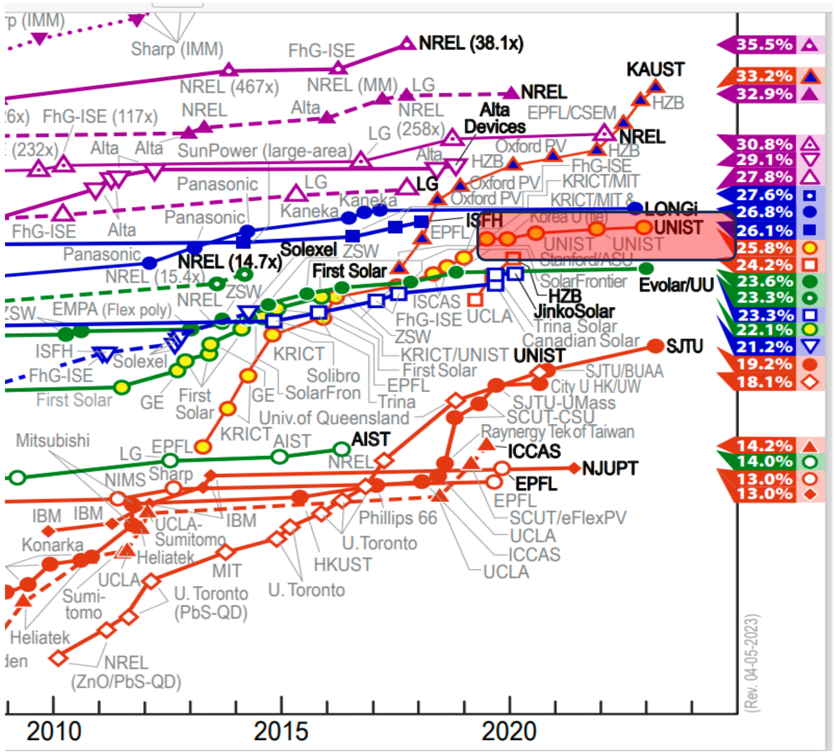


High-performance for

- **Photovoltaics (solar cells)**
- LED, lasing, x-ray detectors, detectors …

Perovskite photovoltaics advance quickly, due to

- Superior optoelectronic properties
- Established theory, technology, materials



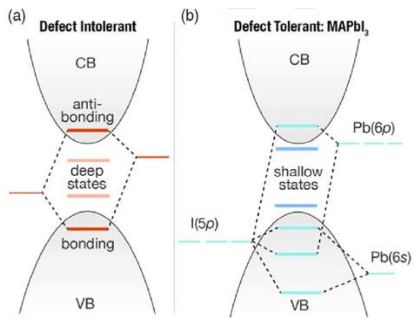
2. Anomalous phenomena: conflict to current semiconductor theory

High-performance of perovskites are usually attributed to superior properties

long carrier lifetime, long diffusion length, defect tolerance, small bonding energy, high absorption, … ??? ***Not sufficient! Not provide physics!***

What makes perovskites so different?

What is physical origin for MHPs' unique optoelectronic properties?



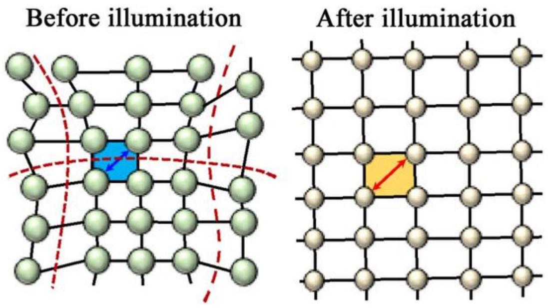
Long carrier lifetime ! ?

1. Much longer than prediction by Langevin theory
2. Significantly extension under continuous excitation
3. Relevant to the dynamic range of system

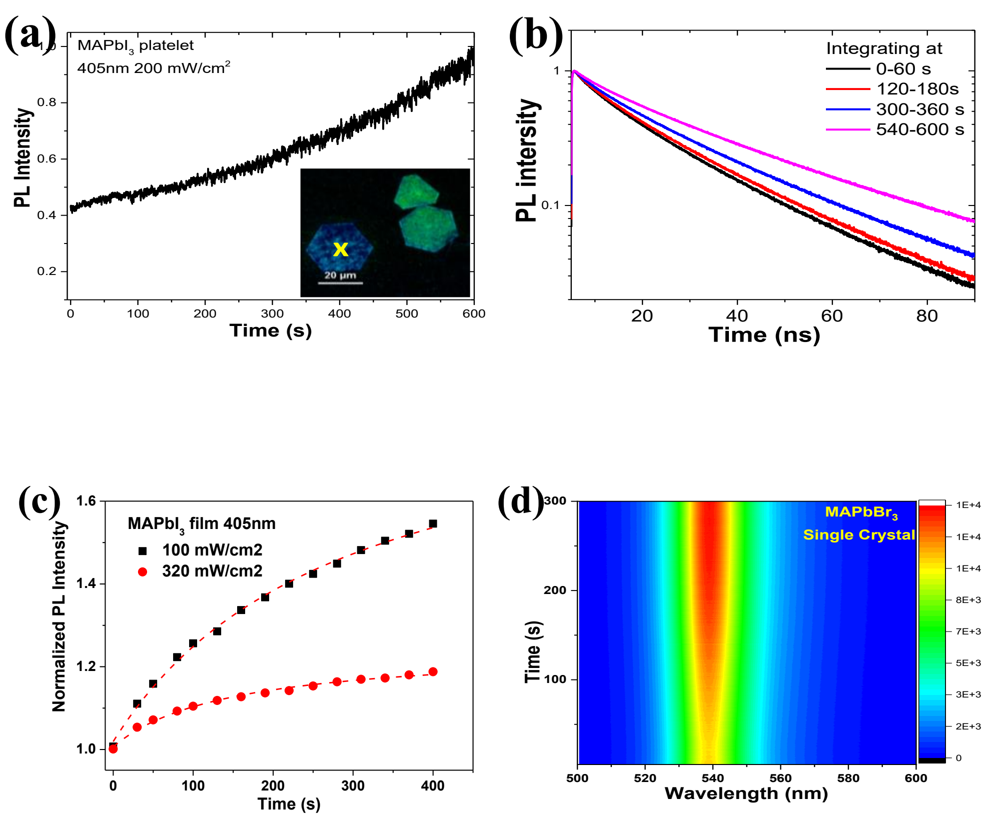
Previously proposed origins for long carrier lifetime/diffusion length **:**

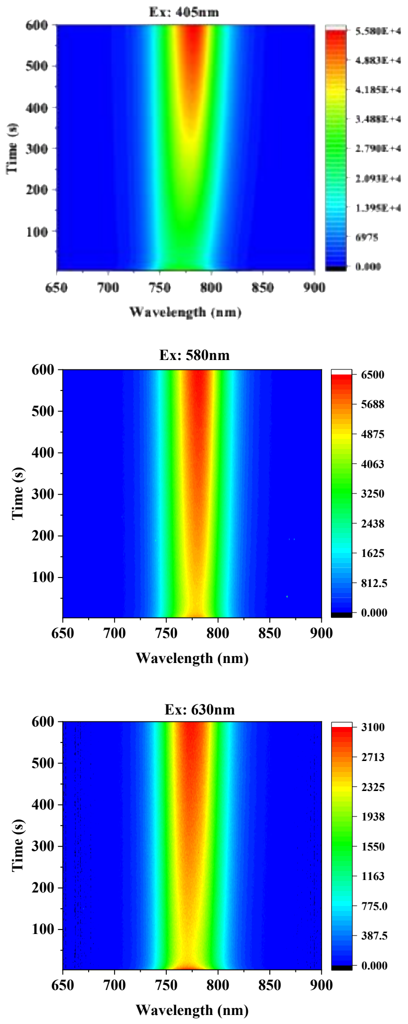
- delayed fluorescence or indirect bandgap transition
- formation of polaron and thus screening
- giant Rashba effect
- ferroelectric domains
- photon recycling

Obviously contradict to some observations → No consensus



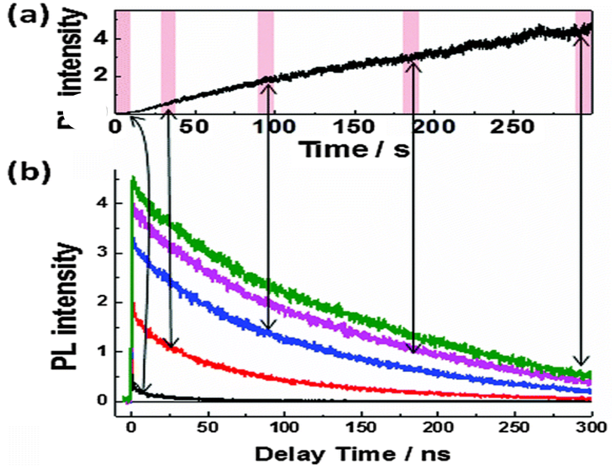
illumination-induced fluorescence enhancement
(photobrightening, defect curing/defect healing)





Under continuous illumination PL efficiency increases and carrier lifetime increases

This is enhancement and reversible, excluding degradation /decomposition



Ultraslow response in MHPs

Ultraslow response ➔ relevant to lattice/strain and/or mobile ions

➢ **Hilde perovskites are soft lattice and exhibit mixed ionic-electronic conduction**

- I-V Hysteresis
- Phase segregation
- Fluorescence intermittency (blinking)
- Fluorescence quenching

mobile ions ➔ detrimental
(charge and defect effects)

- illumination-induced PL enhancement (photobrightening, defect curing/defect healing)
- Defect tolerance
- Memory effect (memoirist)
- upconversion fluorescence
- Persistent structure polarization….

Lattice ➔ beneficial
(lattice, strain, polaron)

Many effects stay in phenomenal description

Defect tolerance / dynamic defect tolerance

long and variable carrier lifetime

photobrightening, defect curing/defect healing

Persistent structure polarization

Memory effect (memoirist)

highly efficient single photon upconversion fluorescence

Illumination induced transparency ……

Conflict to current theory!

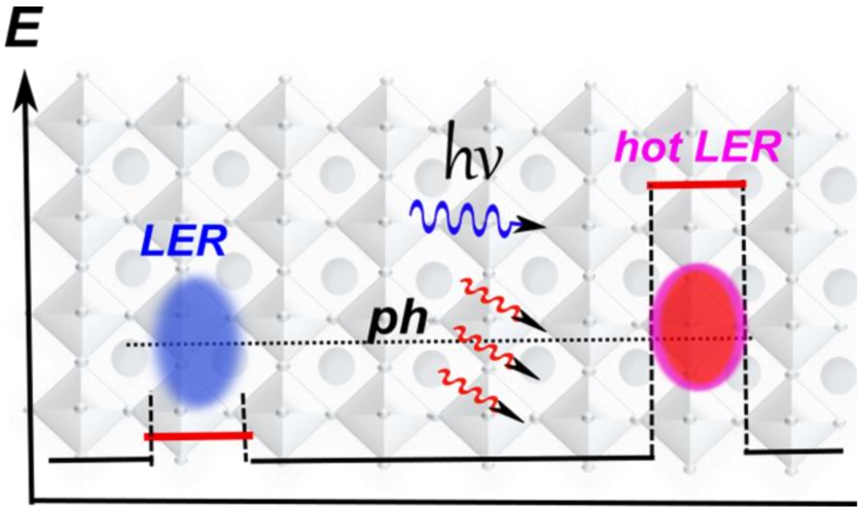
3. *Lattice energy reservoir (LER)*

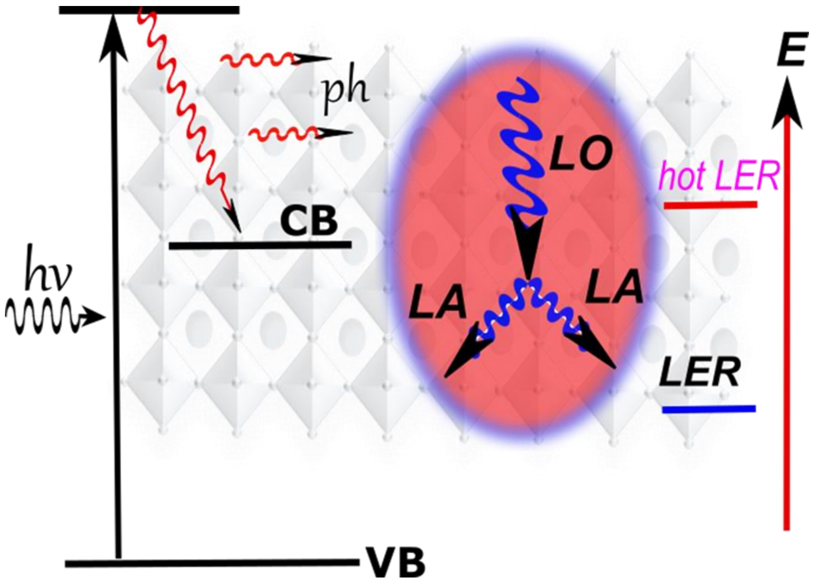
- **Lattice energy reservoir (LER)** are dynamic non-uniformly localized nanodomains. Under light illumination, phonon energy can be stored into LER and generate hot LER (higher potential energy).

- Soft-lattice MHPs can deform/distort lattice thus accumulate energy, forming hot LER.

- LER can form phonon cavity; the strain interface significantly suppress thermal transport/dissipation ➔ **ultralow thermal transport/energy leaking**!

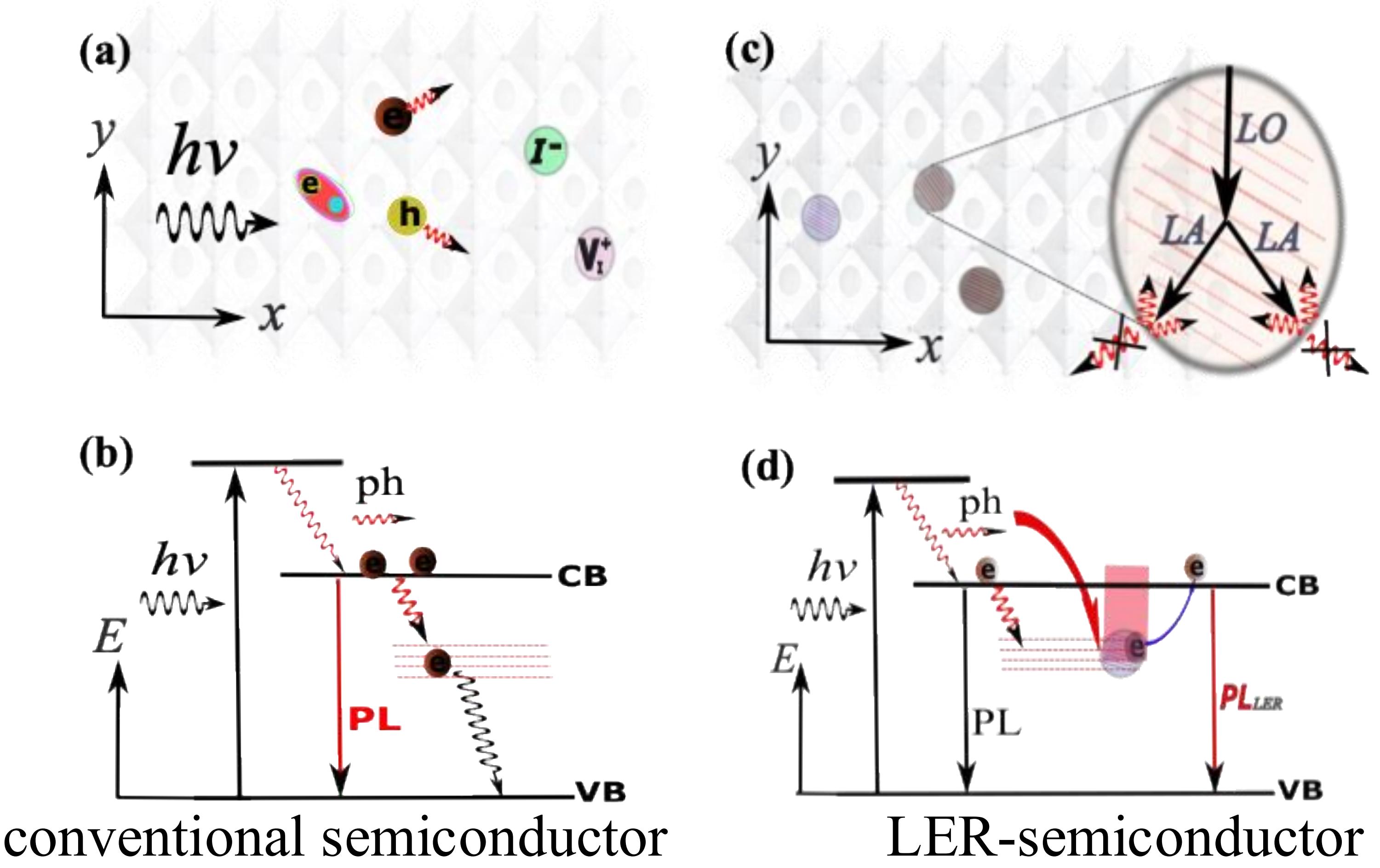
- When subgap electrons drift into the hot LER, they can be upconverted to the conduction band ➔ **upconversion**.





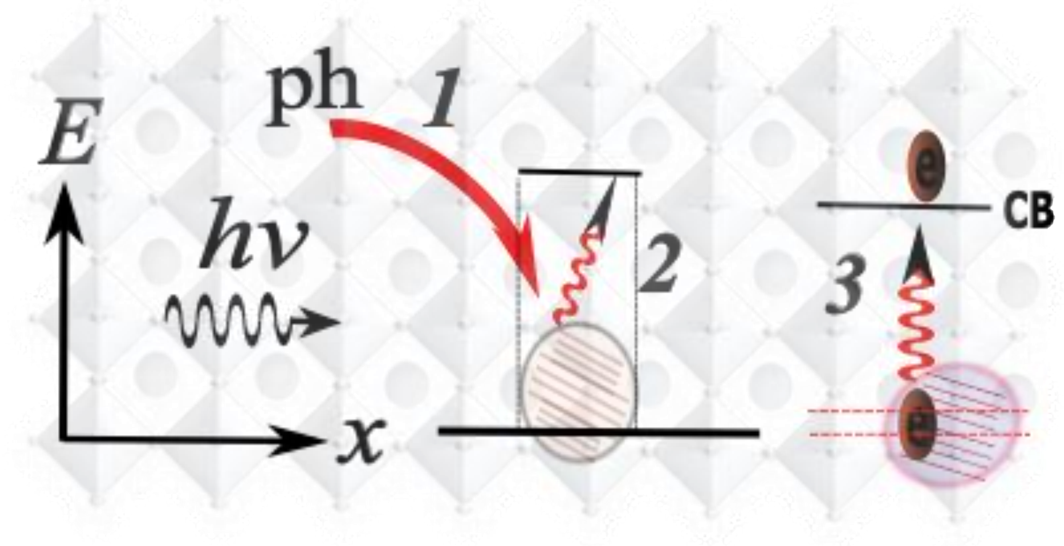
Wen *et al*. Lattice Energy Reservoir in MHPs, ***Account of Materials Research***, 2025 10.1021/accountsmr.5c00047

Carrier dynamics in conventional semiconductor and LER-semiconductor



Energy micro-recycle for defect tolerance:
phonon injection ➔ hot LER formation ➔
subgap carrier upconversion

Energy micro-recycle process in LER:



- 1: phonon energy inject into LER (polaron)
- 2: energy storage as elastic potential energy then hot LER formation as phonon cavity
- 3: subgap carrier upconversion by multi-phonon absorption, driven by hot LER

Hot phonon injection ➔ hot LER formation ➔ subgap carrier upconversion ➔ carrier in CB

LER's physical effects

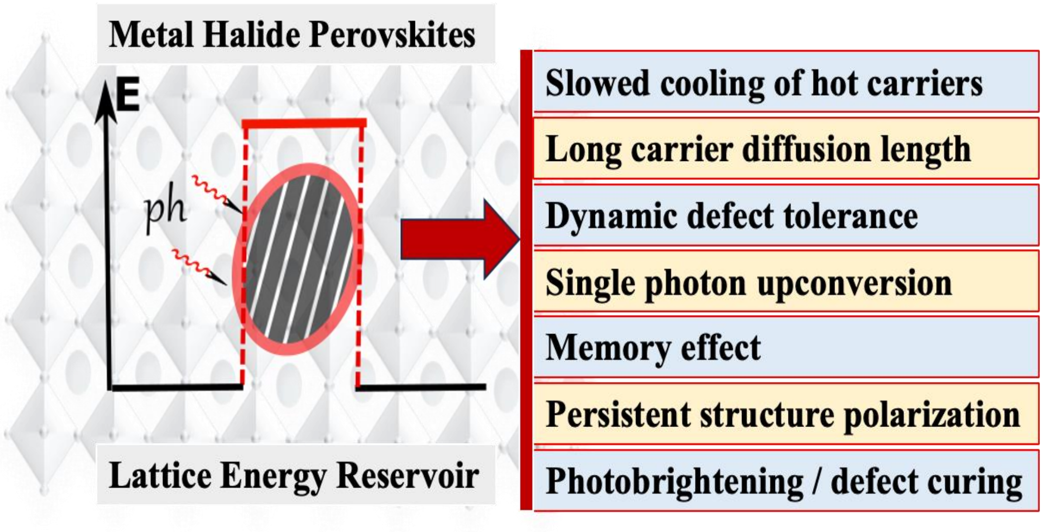
- **slowed cooling by LA→LO upconversion** due to suppressed thermal depletion thus high density LA phonon in LER

- significantly **prolonged carrier lifetime, increased PL efficiency → dynamic defect tolerance**

- **Efficient single photon upconversion fluorescence**

- **ultraslow phenomena (second – hour) due to suppressed energy dissipation**

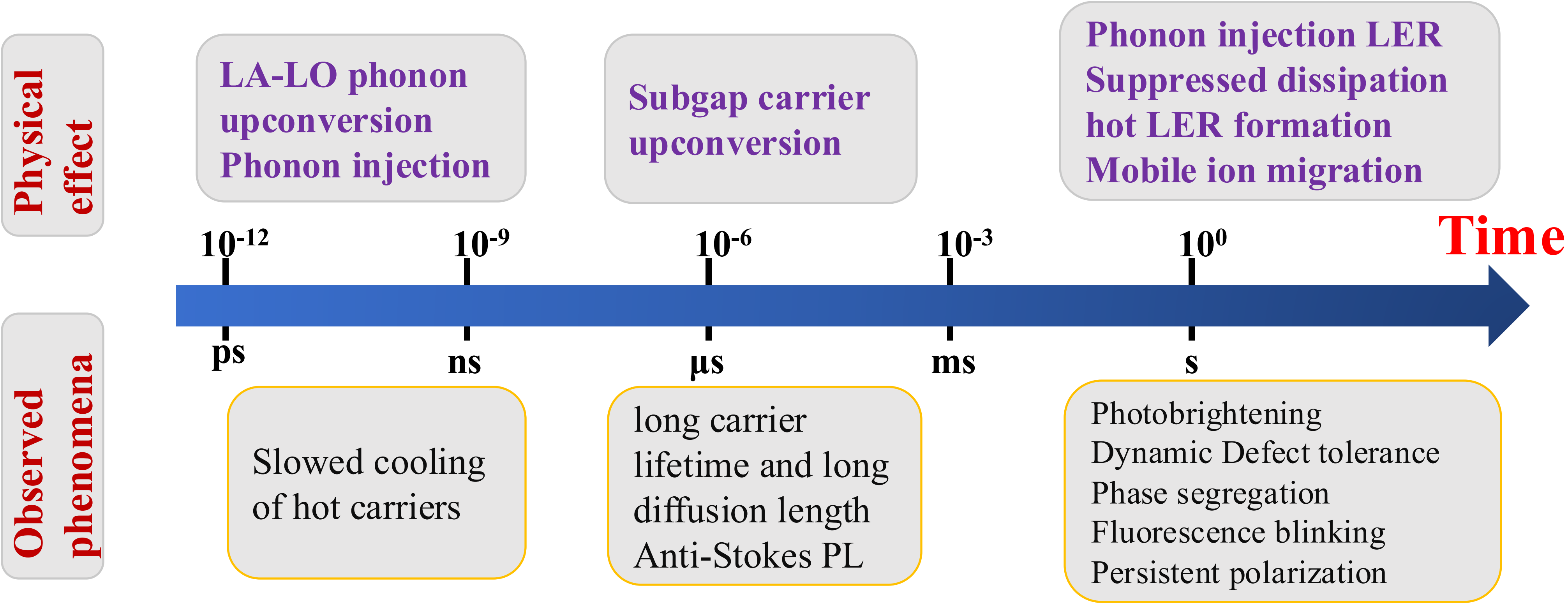
  Persistent structure/lattice polarization
  Memory effect (memoirist)
  Illumination induced transparency ....



→ Fundamental mechanism for unique optoelectronic properties & high-performance devices!

The physical effects of lattice energy

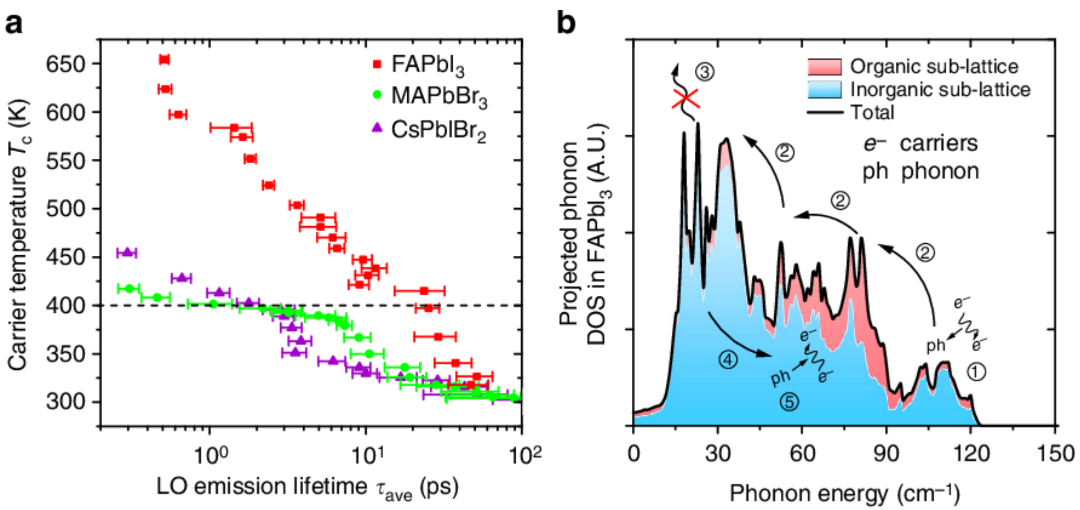
Comprehensively change phonon-carrier-lattice-mobile ion dynamics

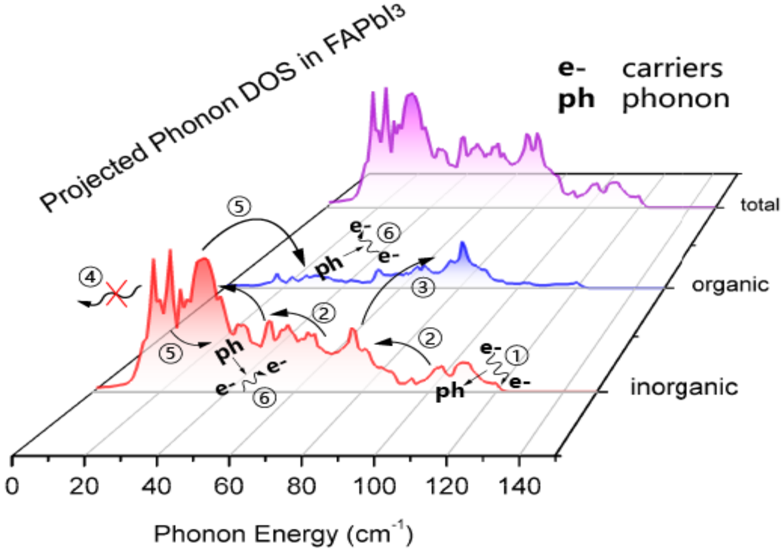


Wen *et al*. Lattice Energy Reservoir in MHPs, ***Account of Materials Research***, 2025

Slowed cooling of hot carriers – phonon bottleneck

Acoustic–optical phonon up-conversion ➔ slowed thermalization





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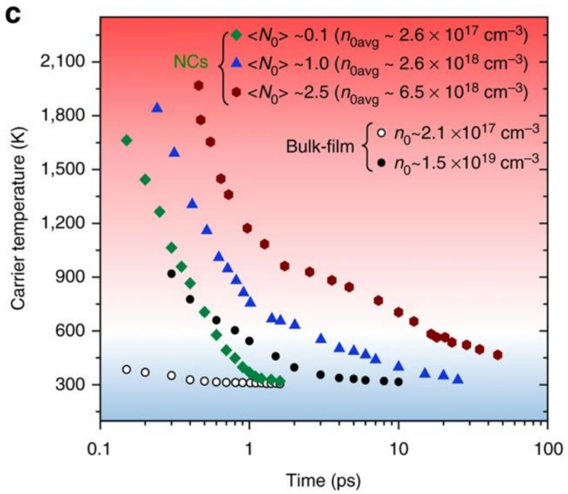
Acoustic-optical phonon up-conversion and hot-phonon bottleneck in lead-halide perovskites

Jianfeng Yang, Xiaoming Wen, Hongze Xia, Rui Sheng, Qingshan Ma, Jincheol Kim, Patrick Tapping, Takaaki Harada, Tak W. Kee, Fuzhi Huang, Yi-Bing Cheng, Martin Green, Anita Ho-Baillie, Shujuan Huang, Santosh Shrestha, Robert Patterson & Gavin Conibeer

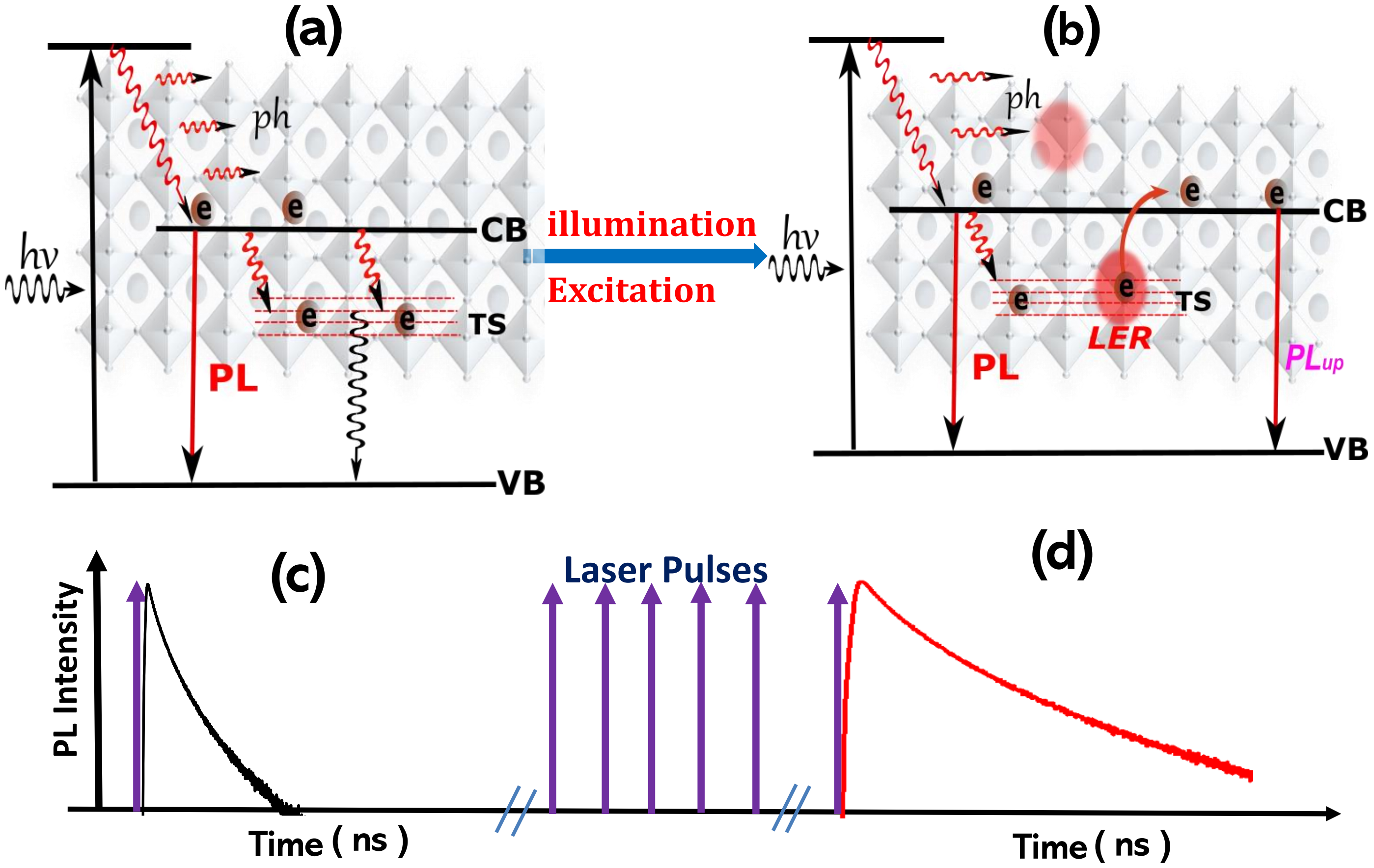


Yang et al. ***Nature Comm***. 8, 14120 (2017)

LER's physical effects

➔ Increased PL efficiency, prolonged carrier lifetime and diffusion length

➔ defect healing/curing, defect tolerance



Key dynamic processes:

- Hot LER formation by lattice-phonon coupling under light illumination
- Subgap carries are upconverted by hot LER

In addition to composition, fabrication quality, excitation parameters, detection system sensitivity also impact the carrier lifetime ➔ NOT intrinsic carrier lifetime!

Measured carrier lifetime is relevant to

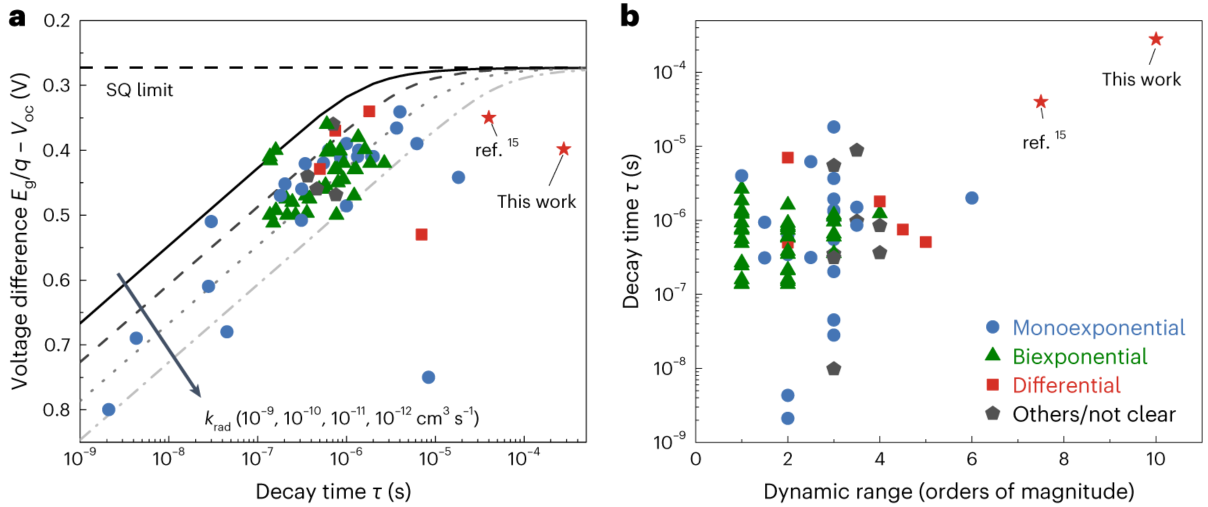
- Measurement parameters: excitation fluence, wavelength
- Dynamic range of detection system （sensitivity）

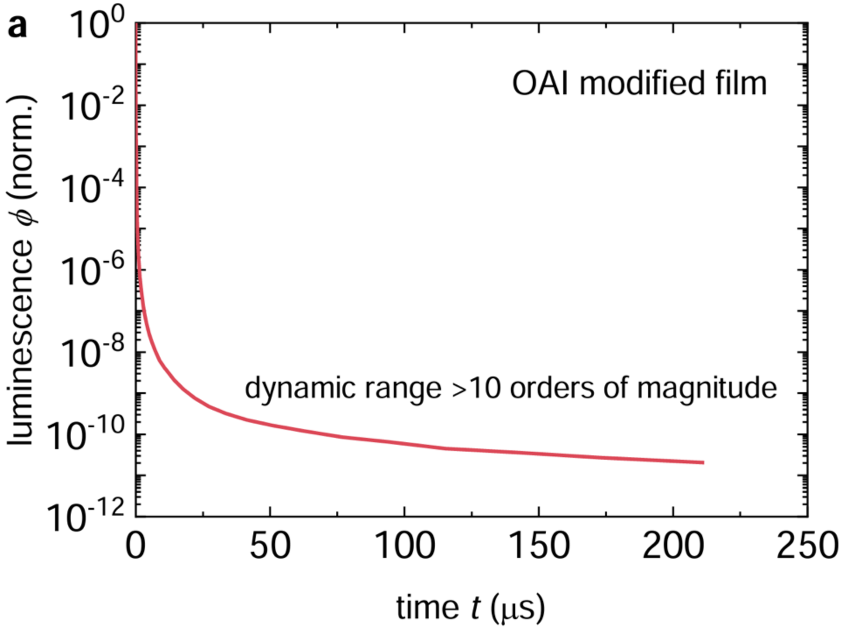
Shallow defects and variable photoluminescence decay times up to 280 μs in triple-cation perovskites

Ye Yuan, Genghua Yan, Chris Dreessen, Toby Rudolph, Markus Hülsbeck, Benjamin Klingebiel, Jiajiu Ye, Uwe Rau & Thomas Kirchartz

Nature Materials 23, 391–397 (2024) | Cite this article

Analysis of reported energy loss and decay time

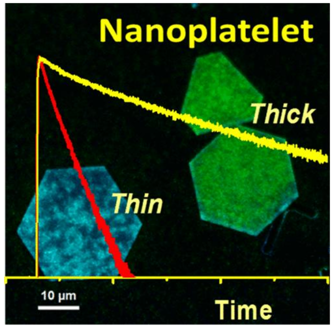




Nature Mater. 23, 391 (2024)

Revisiting charge carrier recombinations

Widely used equation: $$\frac{dn}{dt} = -An - Bn^2 - Cn^3$$



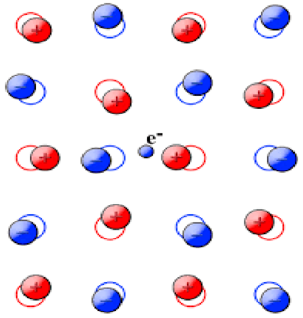
Including SRH, bimolecular and Auger recombinations

Essential assumption:

(a) independent cycles of excitation-dissipation

(b) obtained PL decay curve by integrating many cycles

(c) LER effect (lattice/strain) are ignored

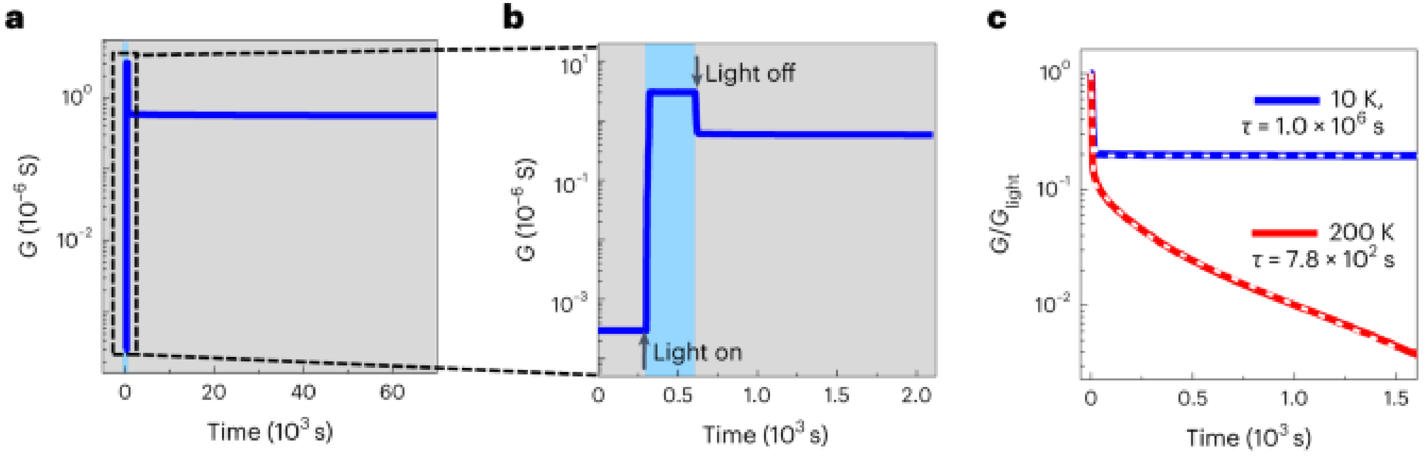


carrier lifetime is also variable during measurement! *What meaning does the measured lifetime represent? It is not intrinsic carrier lifetime!*

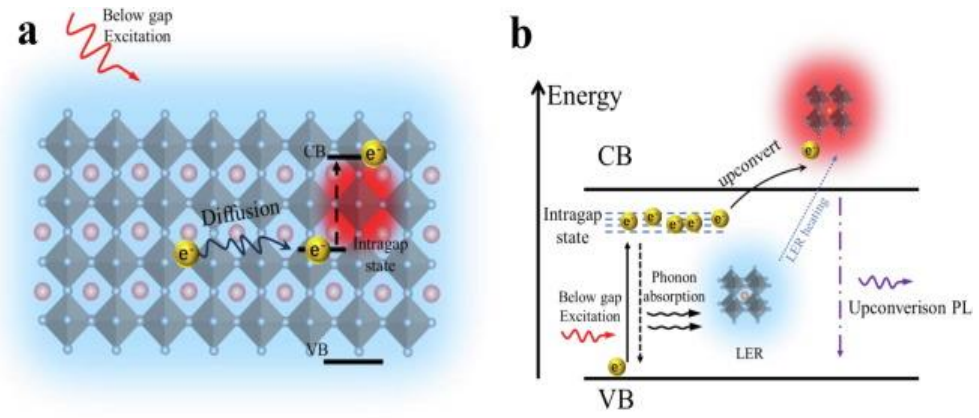
The interpretation for TRPL and TAS needs to re-consider!

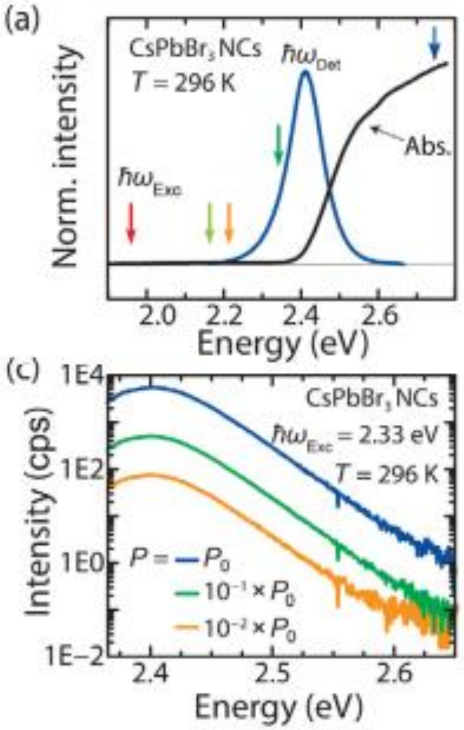
Ultraslow phenomena in MHPs

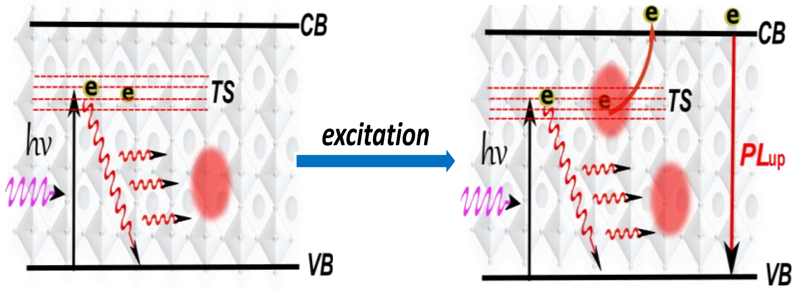
Persistent photocurrent



Efficient anti-Stokes Upconversion PL
(no metal doping)

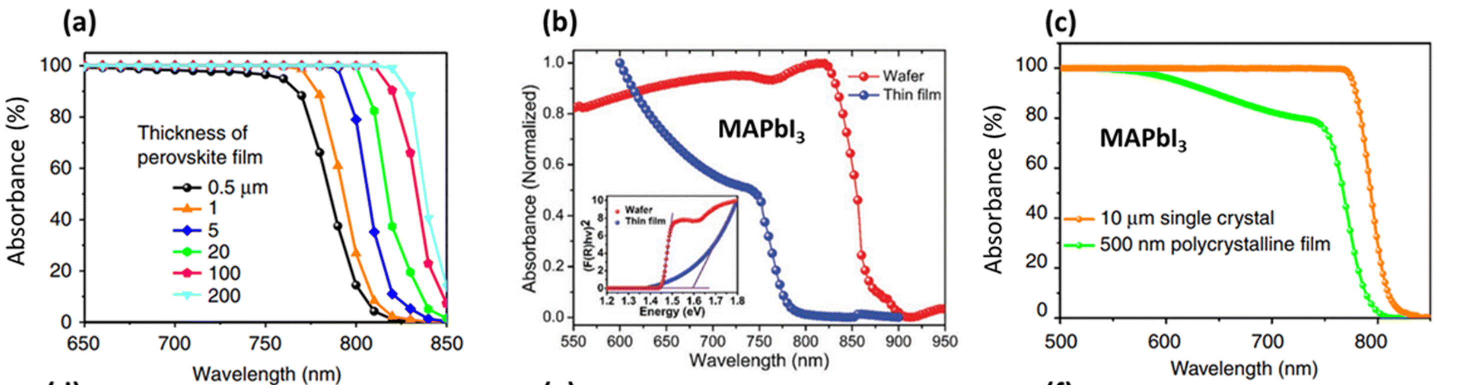


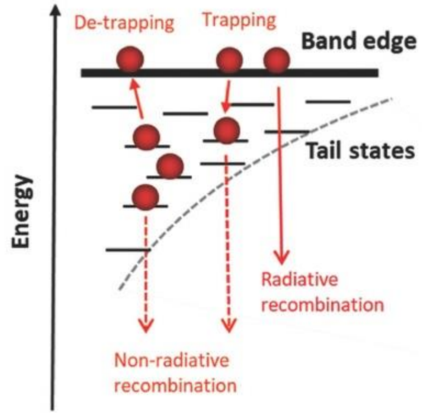




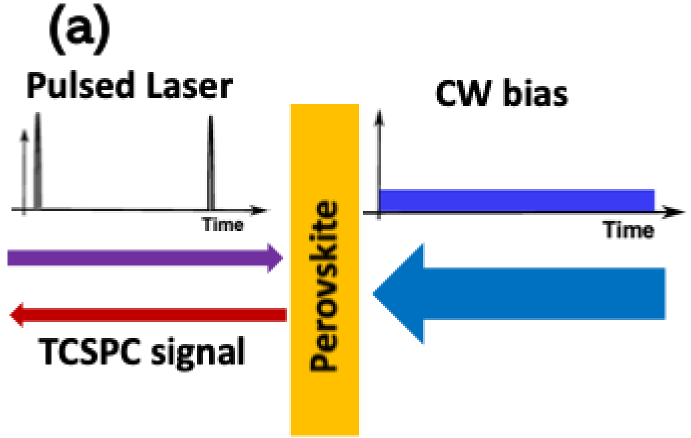
Nature Nanotech. **18**, 357 (2023)
Wen, *et al*. Materials Today 2025

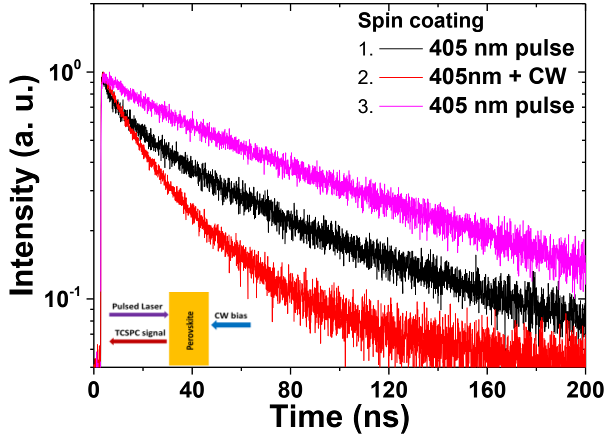
Featured subgap absorption in perovskites





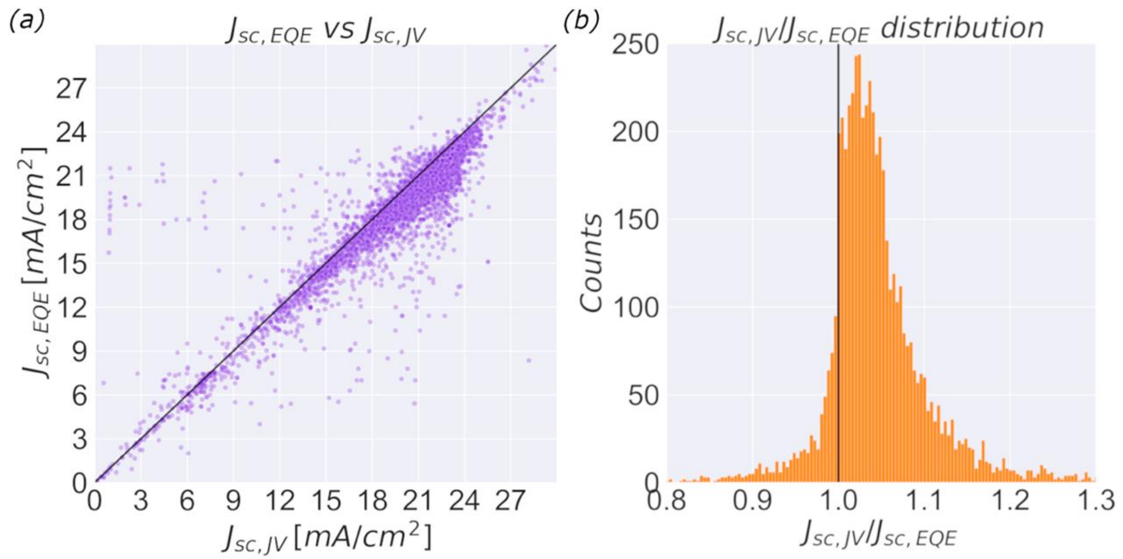
Memory of Carrier lifetime

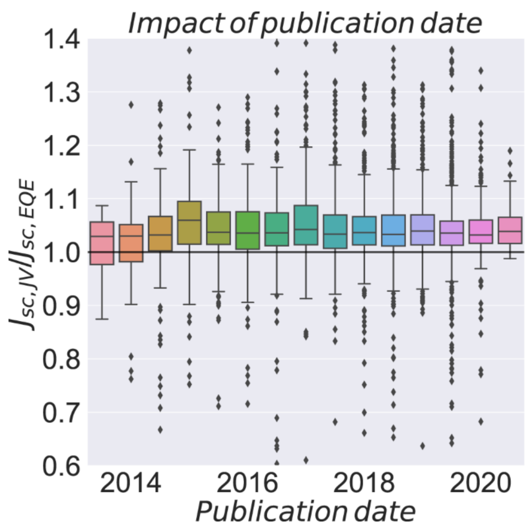




A systematic discrepancy between the short circuit current in perovskite solar cells

$$J_{sc,JV} > J_{sc,EQE} \quad \sim 4\%$$





[Saliba et al. Nature Comm.](#) **14**, 5445 (2023)

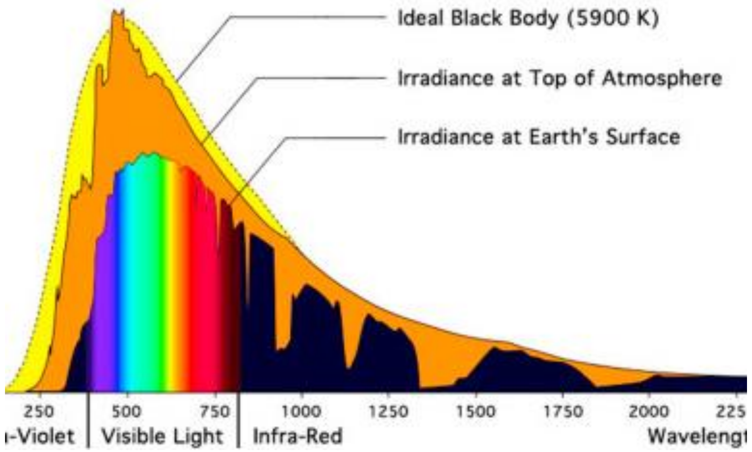
5. *Lattice battery solar cell (LBSC)*

Energy Loss and efficiency limit of solar cells

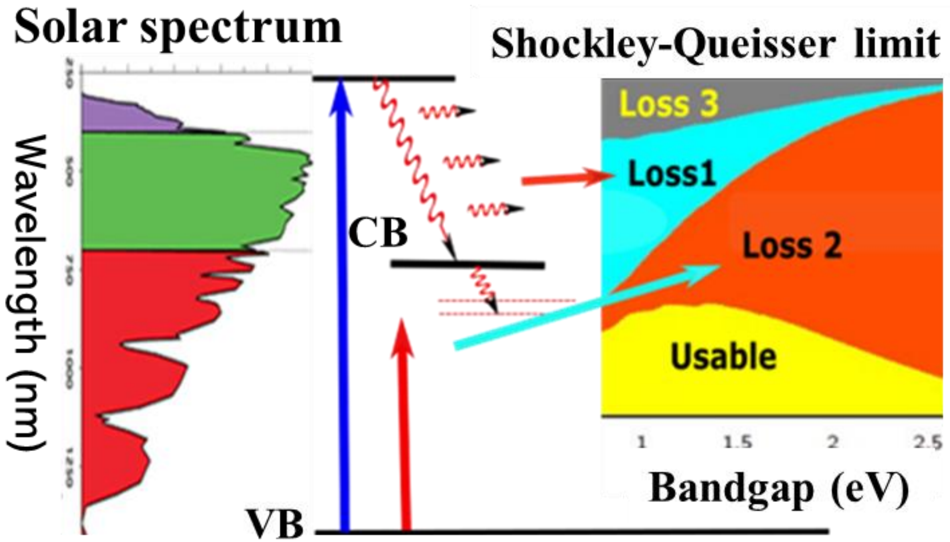
Why > 70% solar energy waste in single junction solar cells:

- ***Loss1: hot phonon from hot carrier***
- ***Loss2: unabsorbed infrared***

➔ efficiency is limited < 33% **Shockley-Queisser limit**



Ultrawide solar spectrum – fixed bandgap

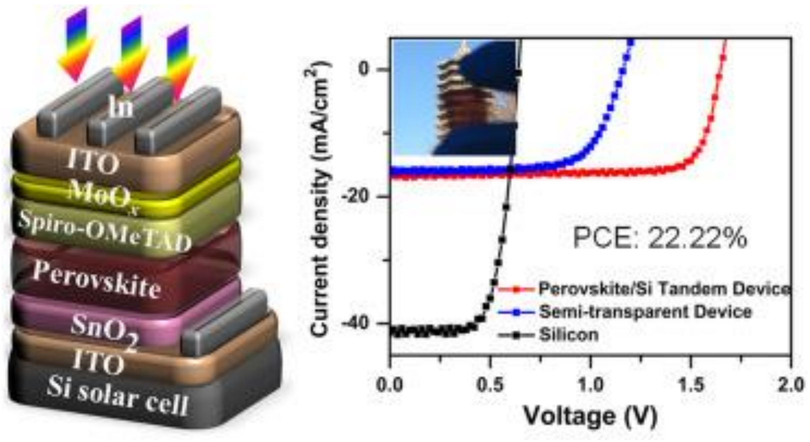


Third generation photovoltaics aim to exceed Shockley-Queisser limit

Previously concepts:

1. Tandem :

Si-perovskite (>33%), perovskite-perovskite (>26%), ..progress recent years, also increase fabrication cost, stability is still challenging



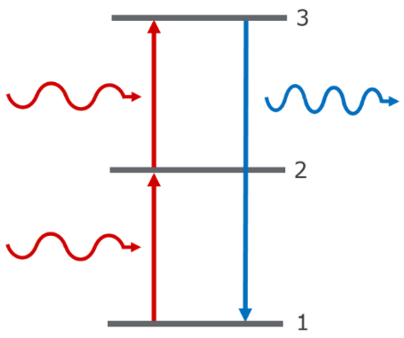
2. Harvesting hot carrier

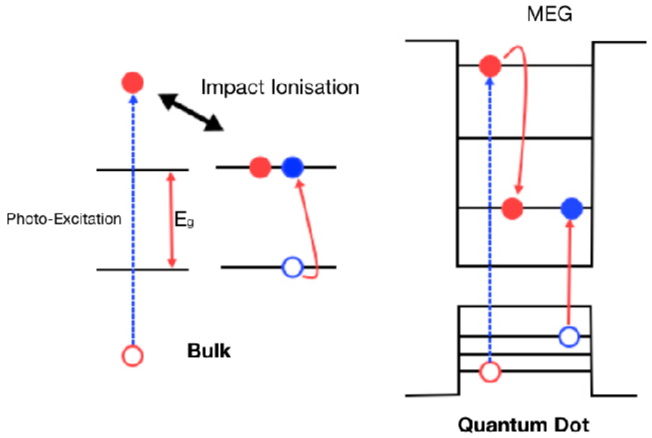
hot carrier solar cells, Multi-exciton generation, singlet fission, (down conversion)

3. Harvesting subgap solar photons

Rare-metal doping upconversion, triplet fusion

Intermediate state



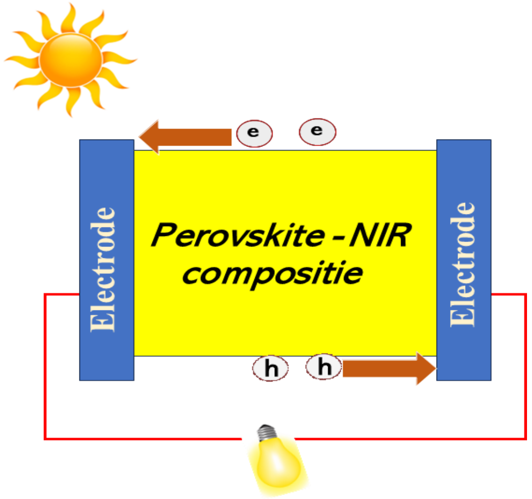


New concept: Lattice battery solar cell (LBSC)

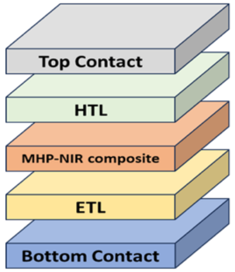
– toward 70% efficiency in single layer

What is Lattice battery solar cell (LBSC)?

synergistic work of lattice energy storage and electron-hole carriers - to achieve wide solar spectrum harvesting (UV-NIR), which can simultaneously overcome the energy losses of hot phonon and sub-bandgap non-absorption in traditional solar cells



- theoretical efficiency of 70% >> SQ limit



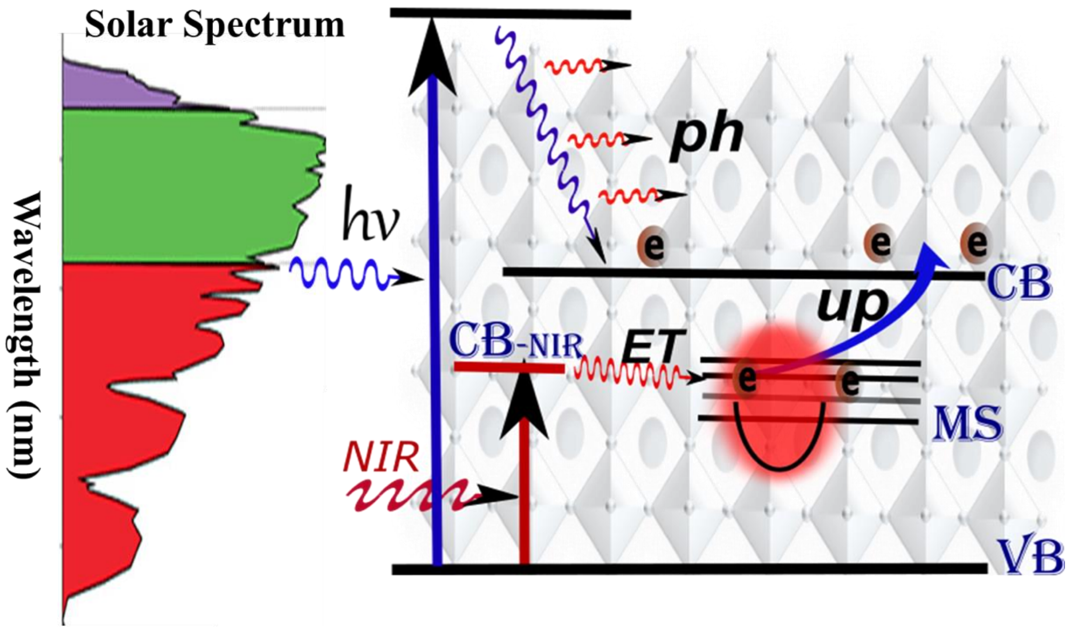
EcoEnergy 2024/ece2.47
The Innovation Energy 2 (2025), 100092

Working principle of LBSC

- **Perovskite-infrared composite** is used as absorber in LBSC for harvesting full solar photon (UV to NIR).
- NIR photons are absorbed by NIR component, photogenerated electrons, then transfer to the subgap state of perovskite and are upconverted to the conduction band
- eventually output as electricity!

Essential difference in solar to electricity:
Other solar cells: solar photon ➔ e, h ➔ electricity
LBSC: solar photon ➔ 1 (e & h ➔ electricity)
➔ 2 (phonon injection and hot LER formation➔ NIR generated carrier transfer to subgap state and upconversion ➔ electricity) ***➔ energy micro-recycle process***

Perovskite-NIR composite



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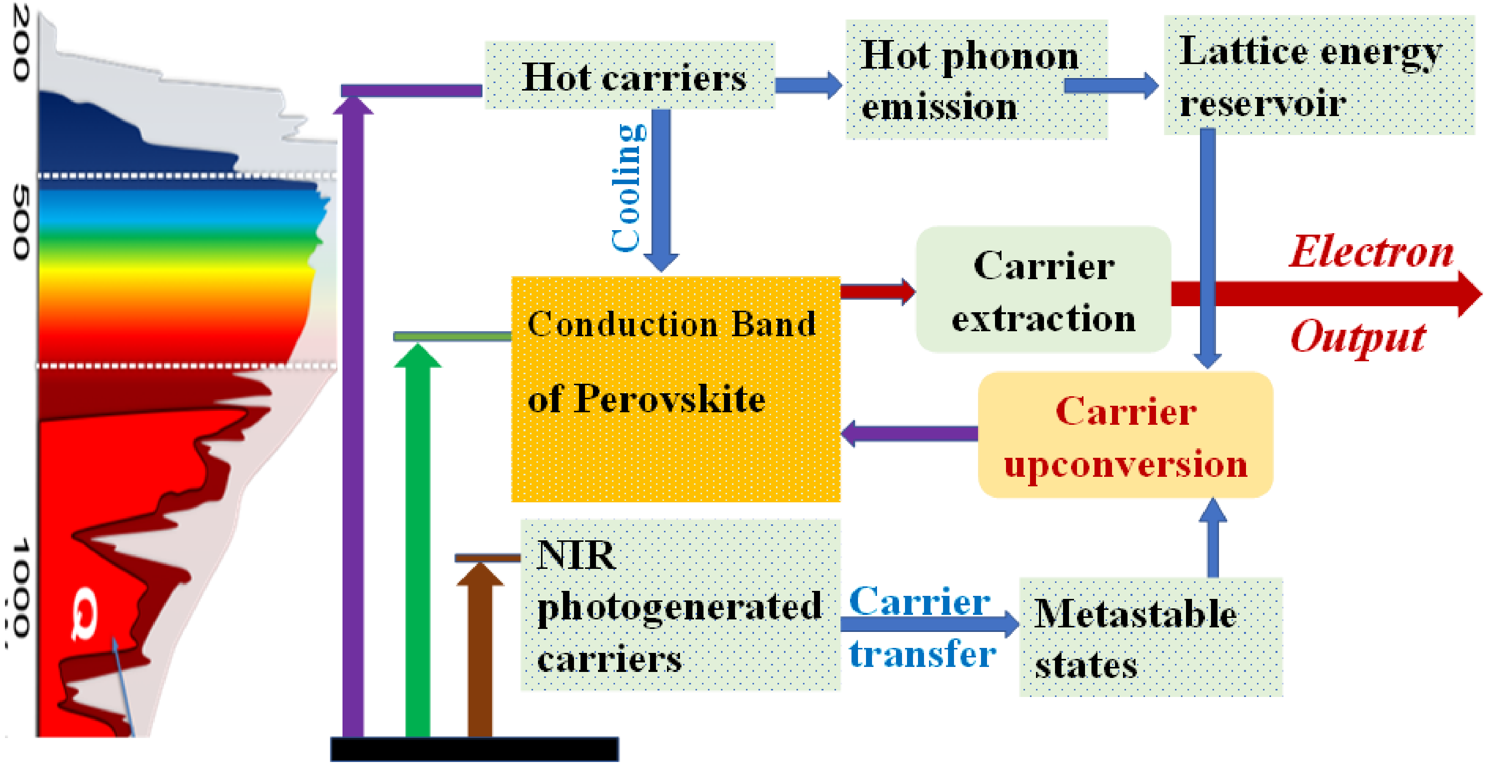
Working processes of LBSC



- **High efficiency:** harvest full spectral solar energy (UV to NIR). Loss1 and Loss2 are converted to electricity

  ➔ **70% efficiency**

- **High stability**: Hot phonon energy is converted into electricity, avoiding heating absorber.

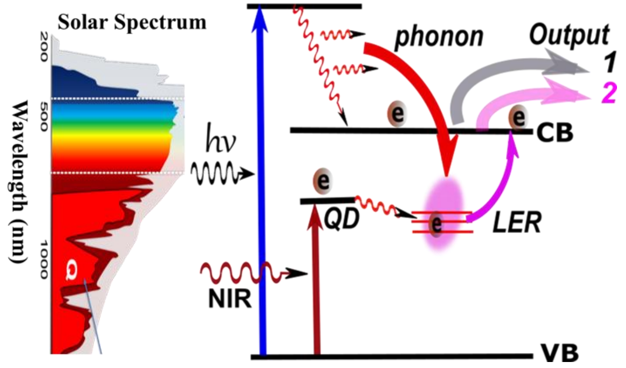
  LBSC will operate at lower temperature, significantly mitigate degradation

Advantages of LBSC

1. high efficiency:

Theoretical efficiency > 70%

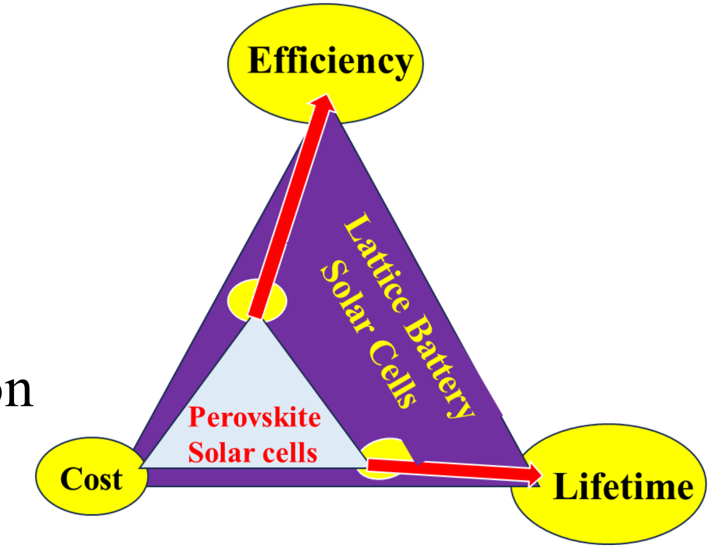
- ➢ Harvesting full spectral solar energy from UV to NIR, typically down to 1 eV (1240 nm)
- ➢ Intrinsically avoid two major losses: hot carrier and sub-bandgap non-absorption



2. high stability

Hot phonon is stored into LER, rather than heating absorber, as electricity output, significantly decrease temperature (actually introduce a cooling mechanism)

3. low-cost fabrication: simple structure of single junction with low-cost solution fabrication

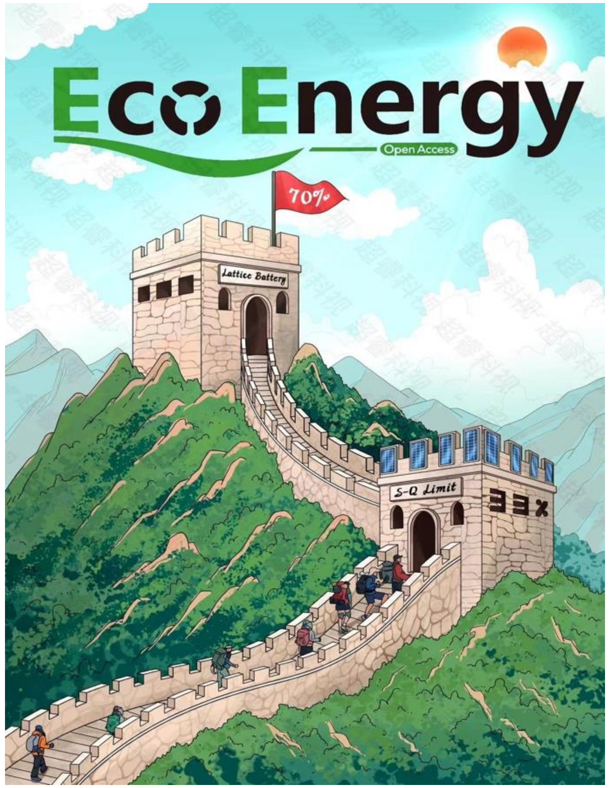


Summary for LER and LBSC

- LER can comprehensively change optoelectronic properties of MHPs ➔ fundamental origin for superior properties. LER define a new type of semiconductors.

- LBSCs operate in a revolutionary mechanism, with high theoretical efficiency, high stability and low-cost fabrication, great promising for next generation solar cells

- A broadband solar energy harvesting strategy-from UV to NIR, promising for solar energy applications: photovoltaics, photocatalysis, self-powered detection, self-powered LED, …



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