# AMANDA
# A Materials Acceleration Platform for Autonomous Solar Cell Optimization

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## Sino-Germany Workshop on Printed Photovoltaics

Erlangen, May 22$^{nd}$ 2024

HI ERN
Helmholtz Institute Erlangen-Nürnberg

part of
JÜLICH Forschungszentrum

in cooperation with
FAU FRIEDRICH-ALEXANDER UNIVERSITÄT ERLANGEN-NÜRNBERG
HZB Helmholtz Zentrum Berlin

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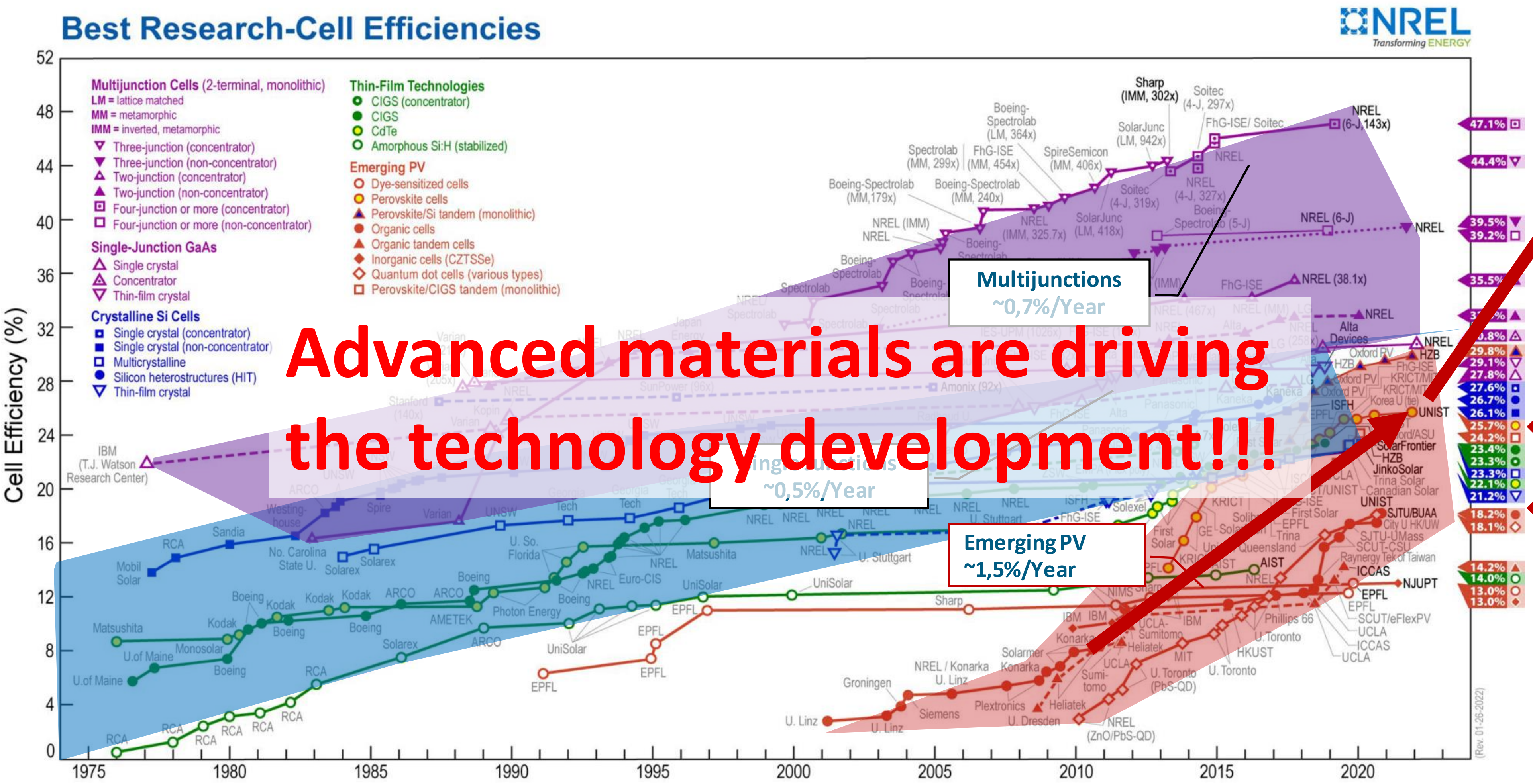
# The development of PV-technologies

## Many technology options for a diversification beyond Silicon




Can we go faster??

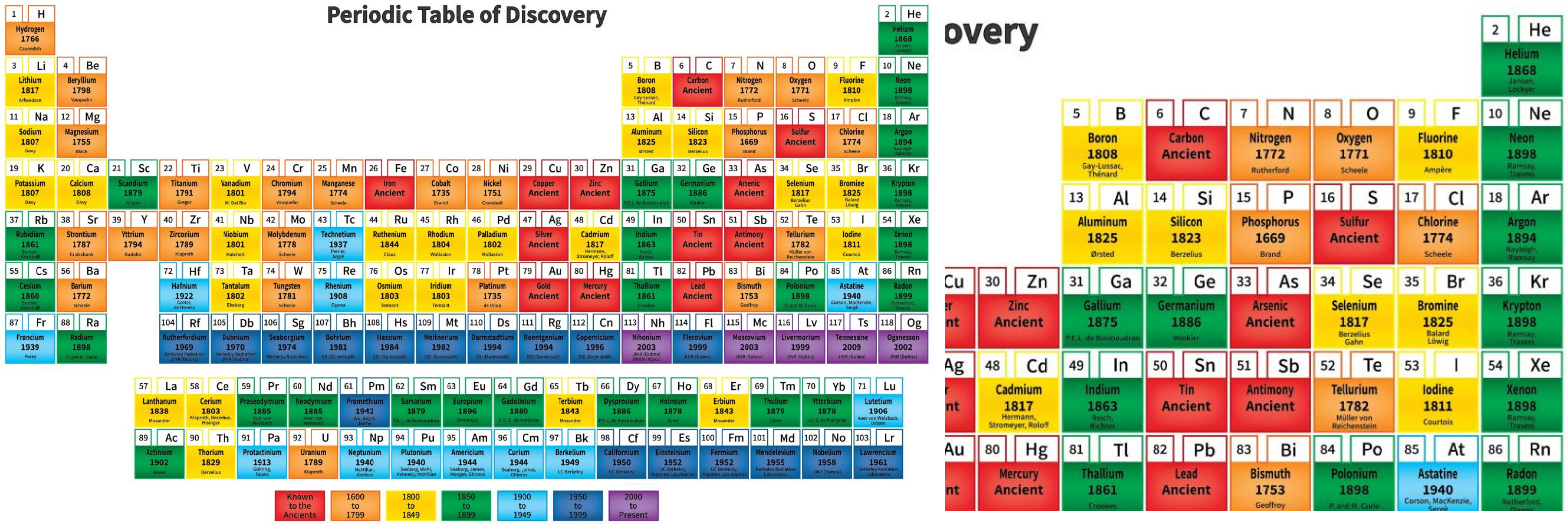
Solution Processed

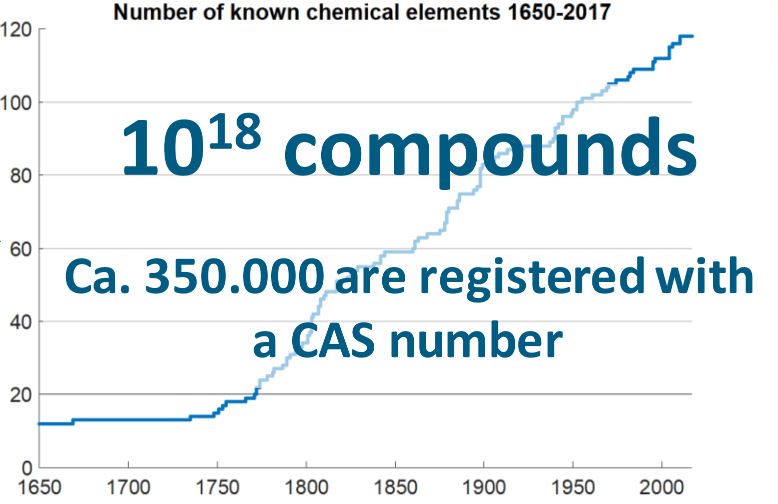
Perovskites (26.1 %)

Organic PV (19.2 %)

NREL, efficiency charts, *https://www.nrel.gov/pv/cell-efficiency.html*

# Complexity at the materials level



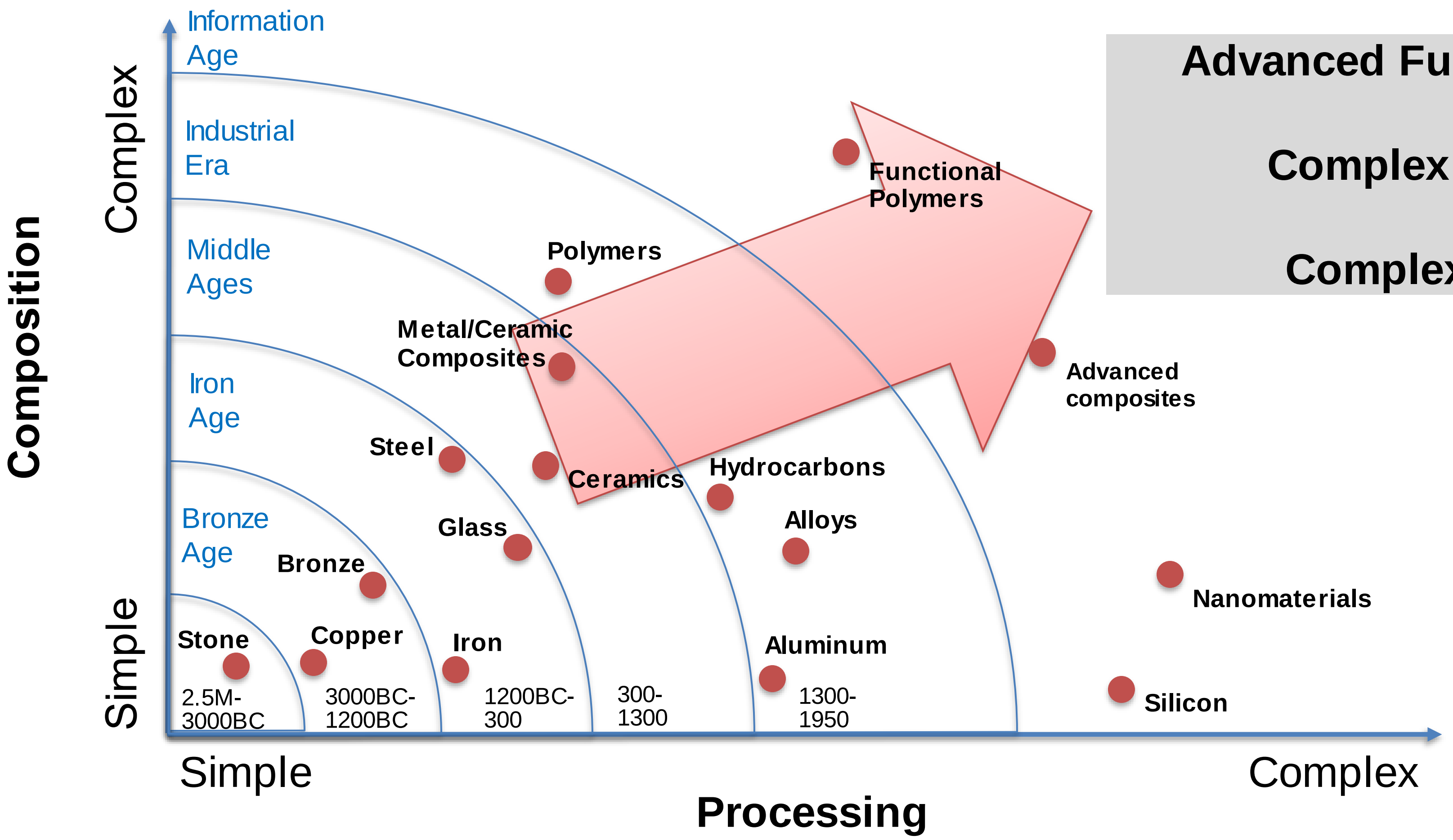




**$10^{18}$ compounds**

**Ca. 350.000 are registered with a CAS number**

Sciencenotes.org: Periodic Table of Discovery, Posted by Anne Helmenstein
Wikipedia.org: Number of known Chemical Elements 1650 to present

# It is not that simple……



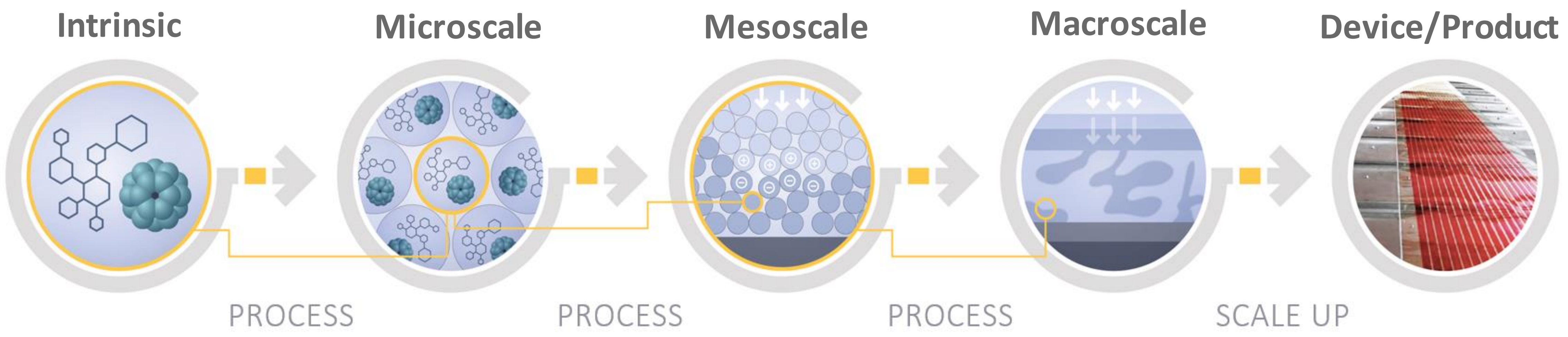

**Advanced Functional Materials**
**=**
**Complex Composition**
**+**
**Complex Processing**

- Materials properties are determined by composition and microstructure
- Microstructure is a result of the processing

Wagner et al., The evolution of Materials Acceleration Platforms: toward the laboratory of the future with AMANDA, Journal of Materials Science (2021), https://doi.org/10.1007/s10853-021-06281-7

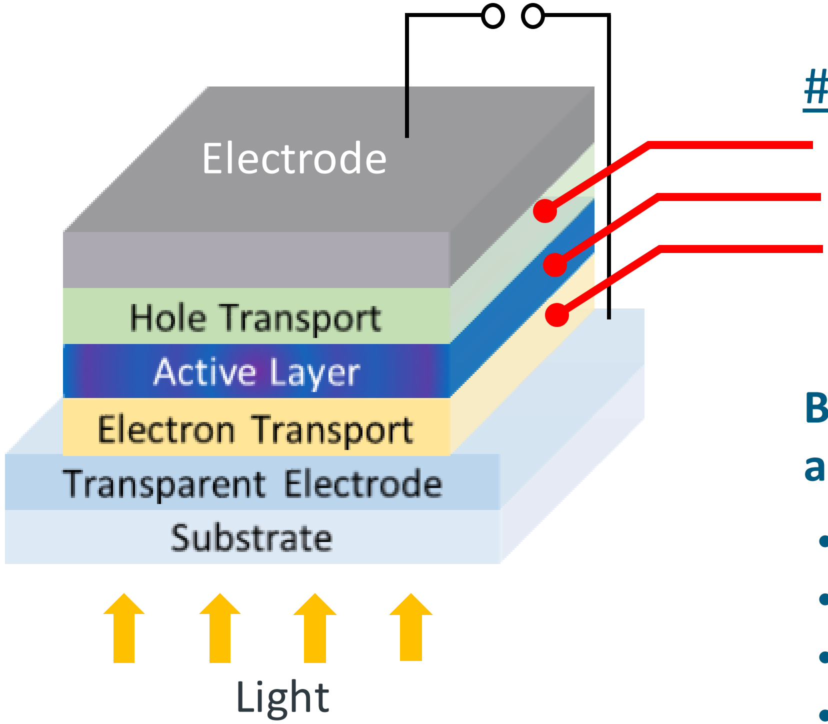
# From Nanoscale to Macroscale

- Materials properties bridge the lengthscales
- Development of functional devices requires global optimization of many parameters (often hidden)




| 10 Materials | 1.000 Composites | 100.000 Layers | 10.000.000 Devices |
| --- | --- | --- | --- |

# A „simple“ solution processed solar cell



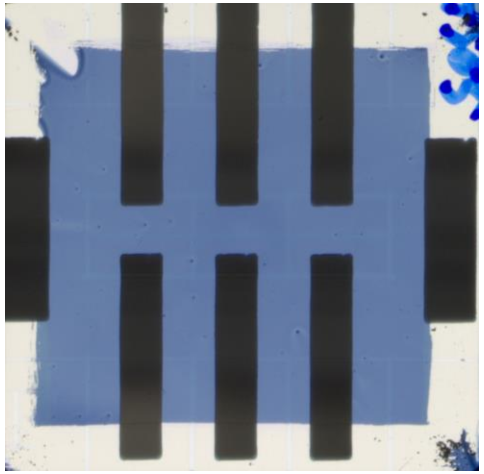

# of options
- ca. $10^1$
- ca. $10^4$ (for a binary mixture)
- ca. $10^1$

ca. $10^6$ in the simplest case!

(For Perovskites $\infty$ possibilities)

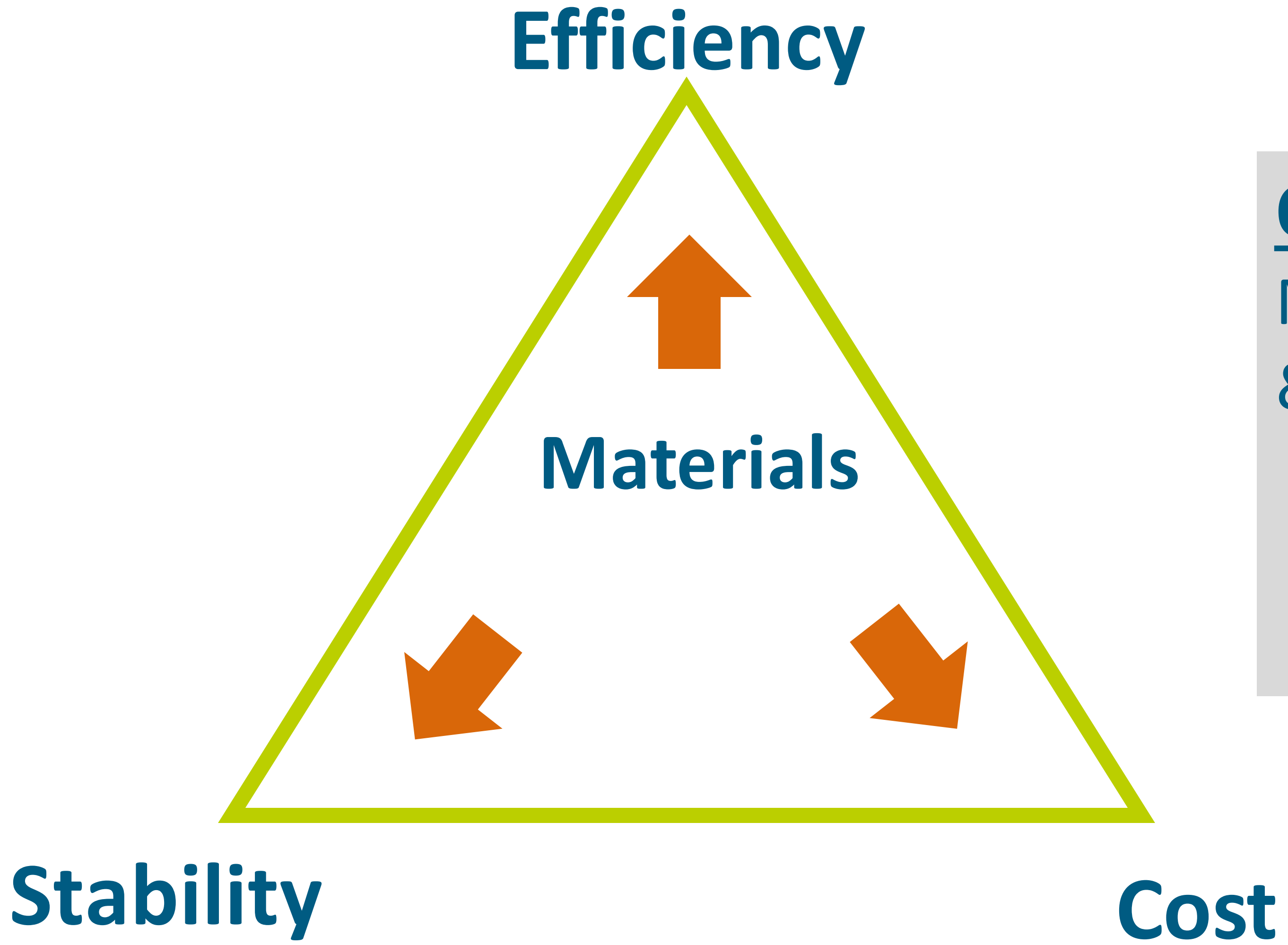
**But we also need to optimize architecture & microstructure:**

- Layer thickness
- Drying speed
- Annealing conditions
- Solvents
- Additives



- **Optimization in a high-dimensional space**
- **Thousands of experiments necessary**

# What does it take for a successful PV-Technology?




**Our Mission**

Massively Parallel Multiparameter & Multiobjective Optimization

- Faster
- With higher precision
- Less material consumption

# HI ERN – High Throughput Methods in Photovoltaics

## Photovoltaics + „Lab 4.0“

### Domain Knowledge

- Physics of Photovoltaics
- Solution processable Semiconductors
- Materials Science & Development
- Semiconductor characterization

### Automation/Data

- Robotics
- Artificial Intelligence & Machine Learning
- Data Bases
- FAIR Data

- Developing approaches to generate high-quality data and evaluate it quickly
- Use machine learning to enable complex optimizations with few experiments

# Automation

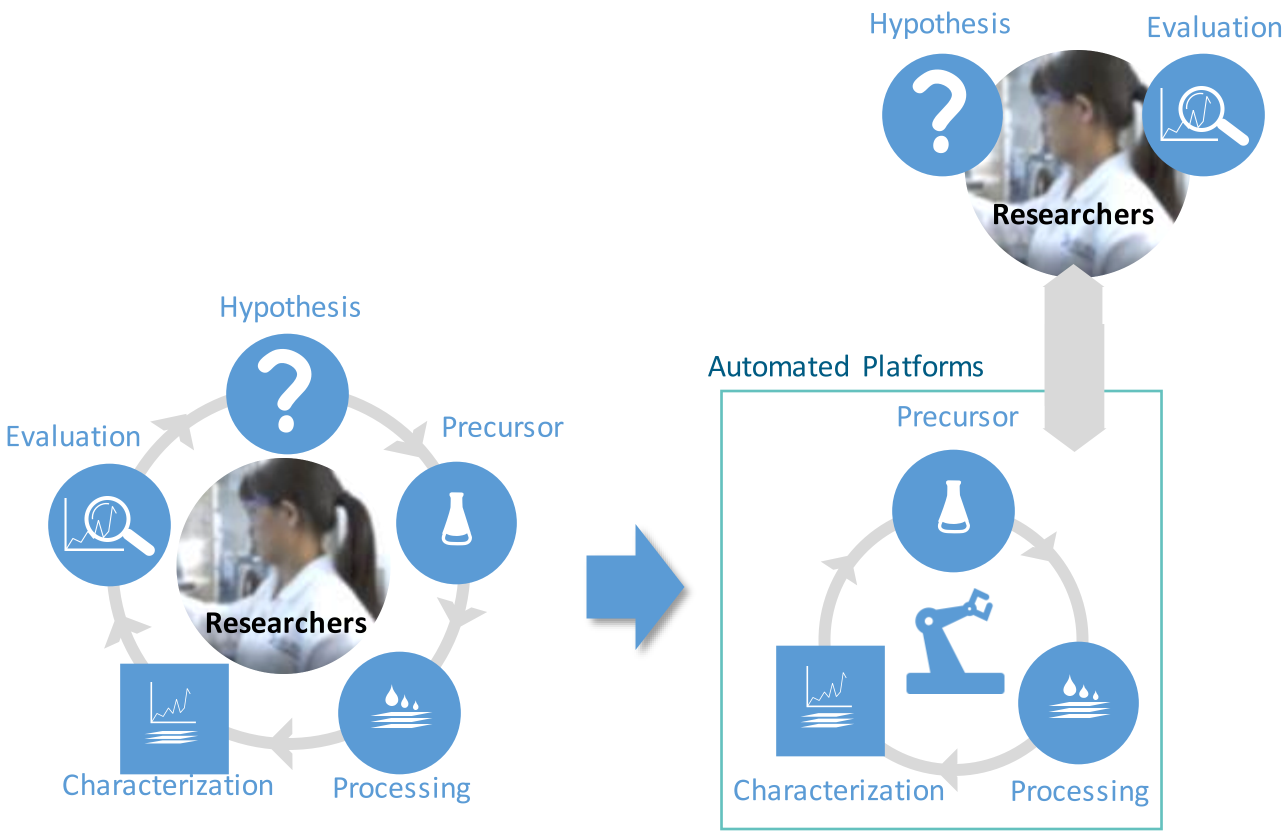
# Materials science today……


**Manual research**

- Personnel intensive
- Highly qualified personnel
- Manual
- Intuitive
- With bad reproducibility
- With terrible transferability

Wagner et al., The evolution of Materials Acceleration Platforms: toward the laboratory of the future with AMANDA, Journal of Materials Science (2021), https://doi.org/10.1007/s10853-021-06281-7

# From Manual to Automated Materials Research




**Manual research**

**Automated research**

- Higher precision
- Higher reproducibility
- Scalability
- Structured data collection
- FAIR data

Wagner et al., The evolution of Materials Acceleration Platforms: toward the laboratory of the future with AMANDA, Journal of Materials Science (2021), https://doi.org/10.1007/s10853-021-06281-7

# Automation of the laboratory – the HI ERN Portfolio

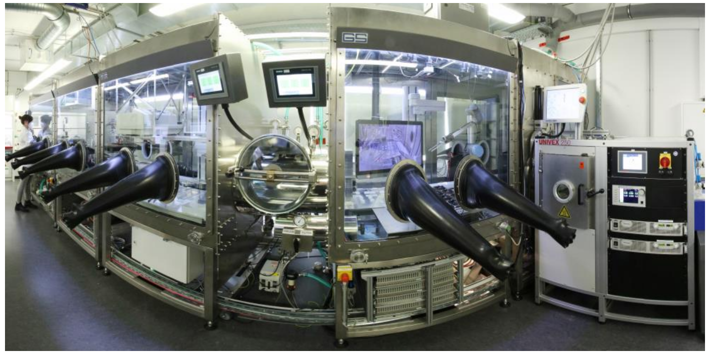
Microwave Reactor

Hamilton Starlet

Tecan Freedom Evo

Spinbot 1 & 2

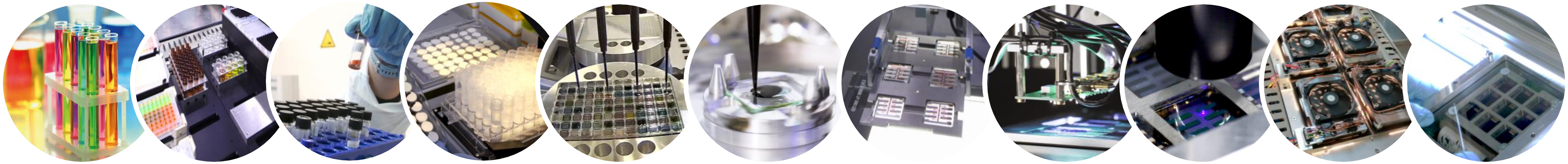
Amanda Line1



X-ALT

**Synthesis** → **Formulation** → **Thin Films** → **Devices** → **Characterization** → **Ageing**

Coupling Reactions | Combinatorial Mixing | Nanoparticle Synthesis | Drop Casting | Spin Coating | Multilayers Metallization | Electrical | Optical | Inline ALT | Offline ALT

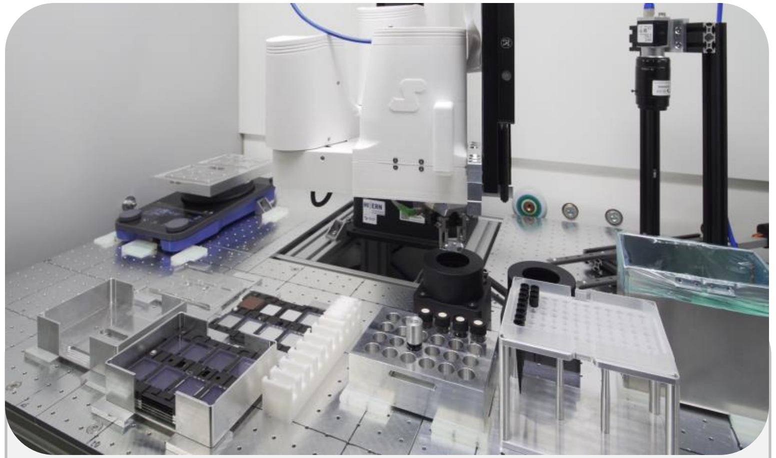


# Equipment for automated testing of materials/devices

## Tecan Freedom Evo

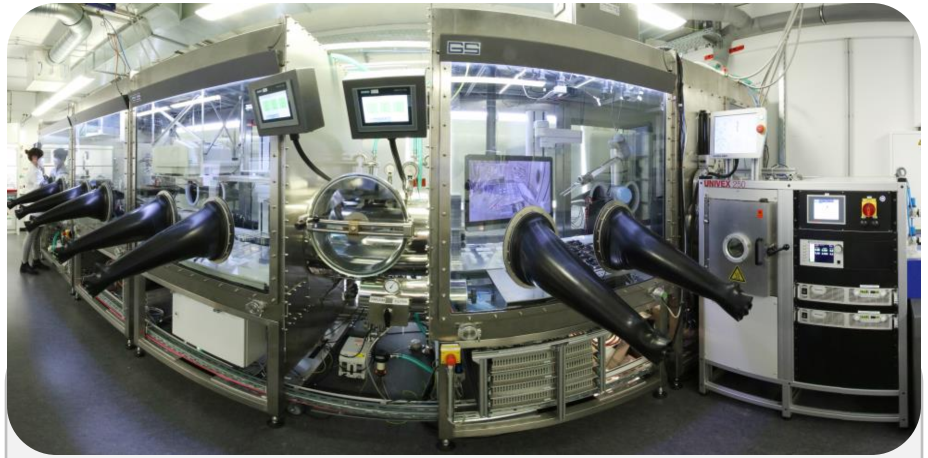

- Pipetting
- Solution formulation
- Dropcasting
- Absorption & PL
- In Air

## SpinBot 1 & 2 & 3



- Pipetting
- Solution formulation
- Dropcasting
- Spincoating/Wiping
- Annealing
- In $N_2$ or Air

## AMANDA-Line1



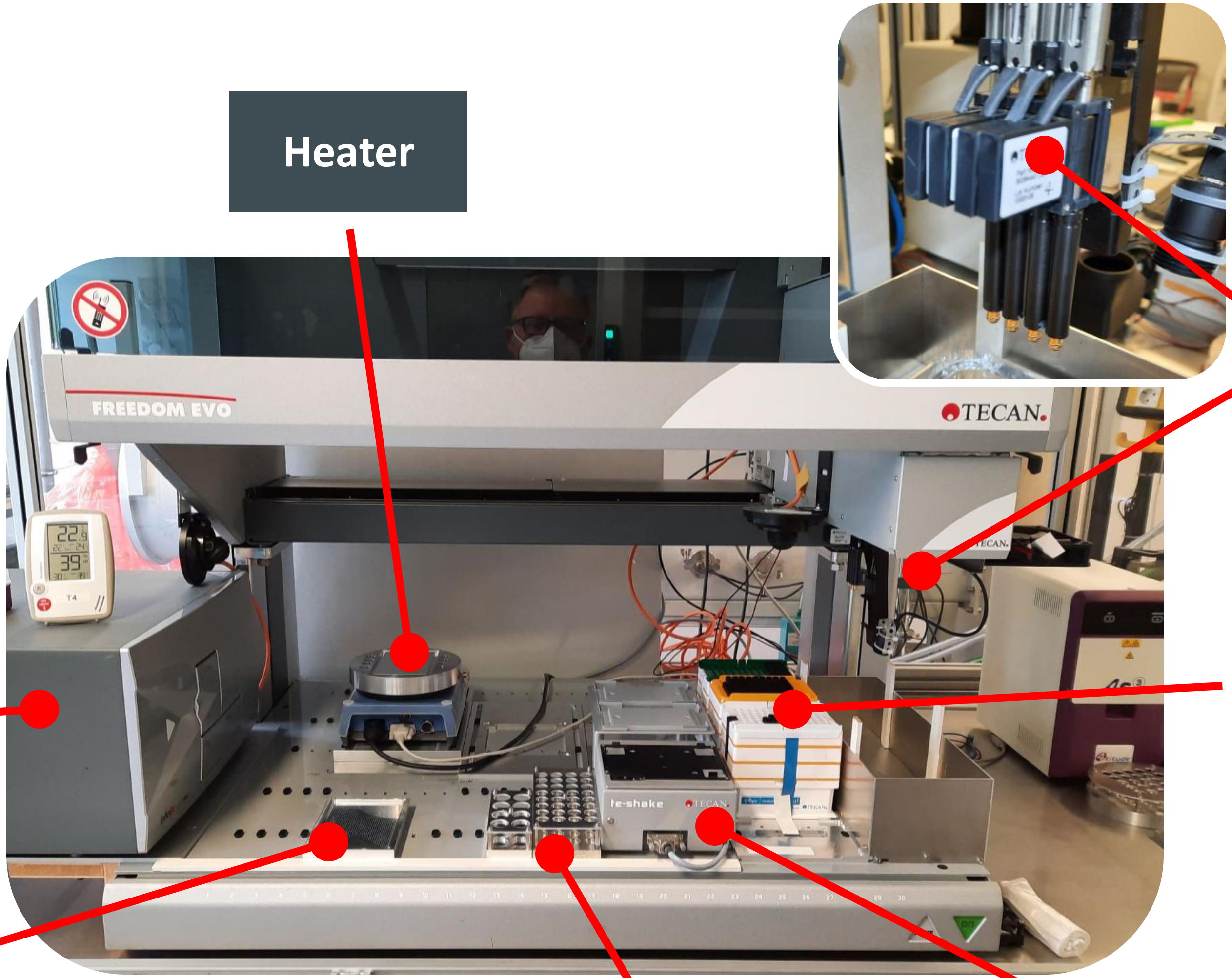
- Pipetting
- Solution Formulation
- Spincoating/Wiping
- Photo
- Absorption & PL
- Evaporation
- jV-Characterization
- Accelerated Aging
- In $N_2$ or Air

➢ Integrated automated Platforms enable HT-research
➢ 5 systems form the core of the Materials and Device work

# The Tecan System - Synthesis and Ink Formulation

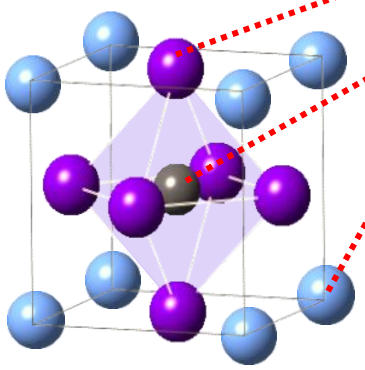
**Capabilities:**

- Pipetting
- Solution formulation
- Dropcasting
- Absorption & PL



# Example Problem: Synthesis of Lead-free Perovskites

## Generic Perovskite: $ABX_3$



X-halide ($I^-$, $Br^-$, $Cl^-$)

B-Cation ($Pb^{2+}$, $Sn^{2+}$)

A-cation

- $Cs^+$
- $Rb^+$
- $K^+$
- $CH_3$-$NH_3^+$
- $NH_2$=CH-$NH_2^+$

**Opportunity:**

- PSKs are a whole class of semiconductors
- Very high PV-efficiencies observed
- Many substitutions possible

**Problem:**

- Record efficiency and stable PSK-cells contain Lead
- Replacement of Lead is non-trivial
- Many combinations possible

## Solution Approach: CANBIC PSKs

$(CsPb^{2+}Cl_3)_2$ $A^IM^{II}X_2$

↓ $2M^{II}$ for $M^IM^{III}$ substitution

$Cs_2(Ag^+Bi^{3+})Cl_6$

↓ isomorphic substitution in $M^I/M^{III}$ positions

$Cs_2(Ag_x + Na_{1-x})(Bi_y + In_{1-y})Cl_6$

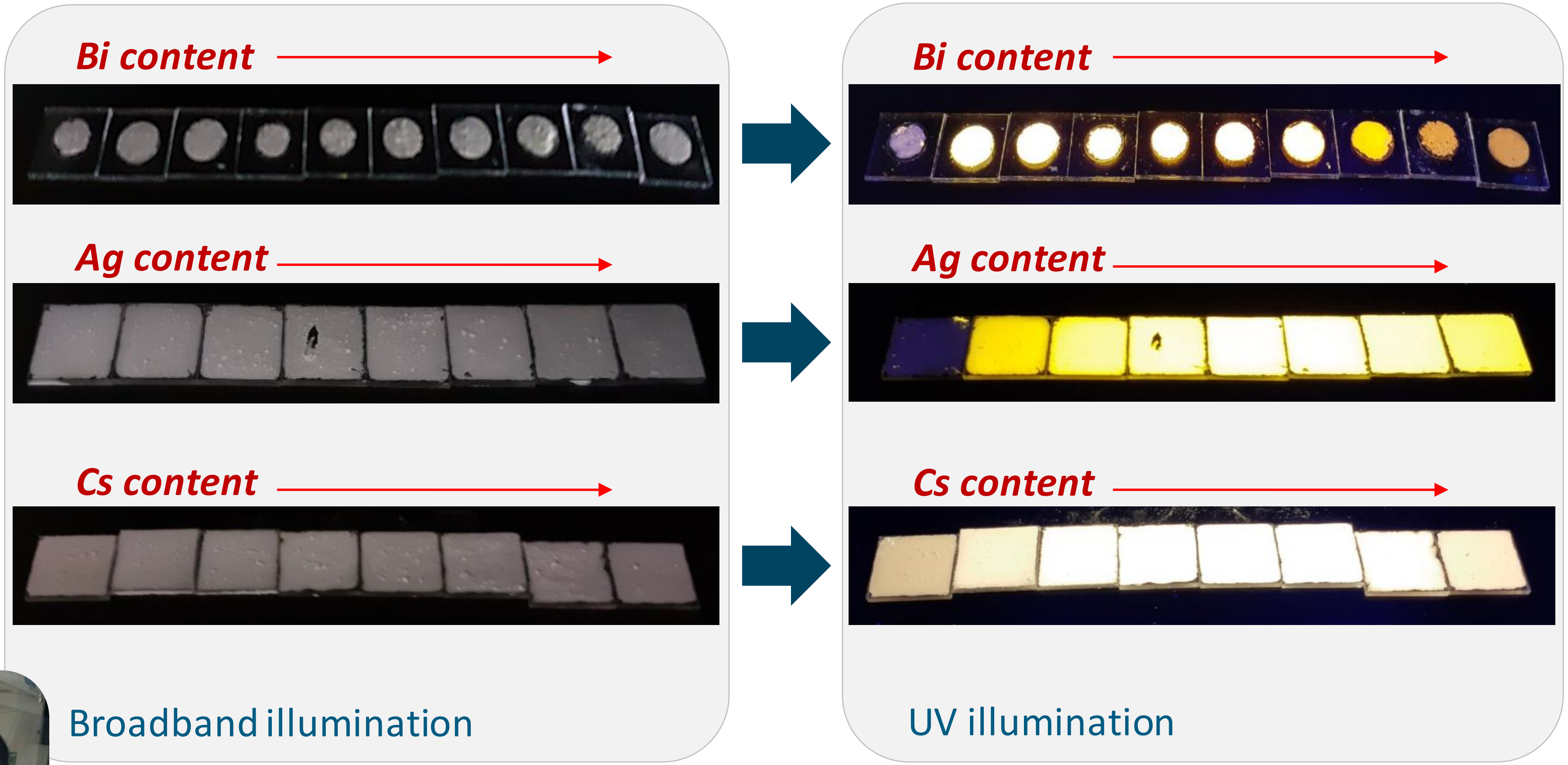
- High photoluminescence (PL) quantum yield (QY)
- Highly stable
- Broad variation of composition in $A^I$ and $A^{III}$ positions
- Synthesis in ambient conditions

# Manual Exploration of materials space

$Cs_2(Ag,Na)Bi_yIn_{1-y}Cl_6$

$Cs_2Ag_xNa_{1-x}(Bi,In)Cl_6$

$Cs_z(Ag,Na)(Bi,In)Cl_6$



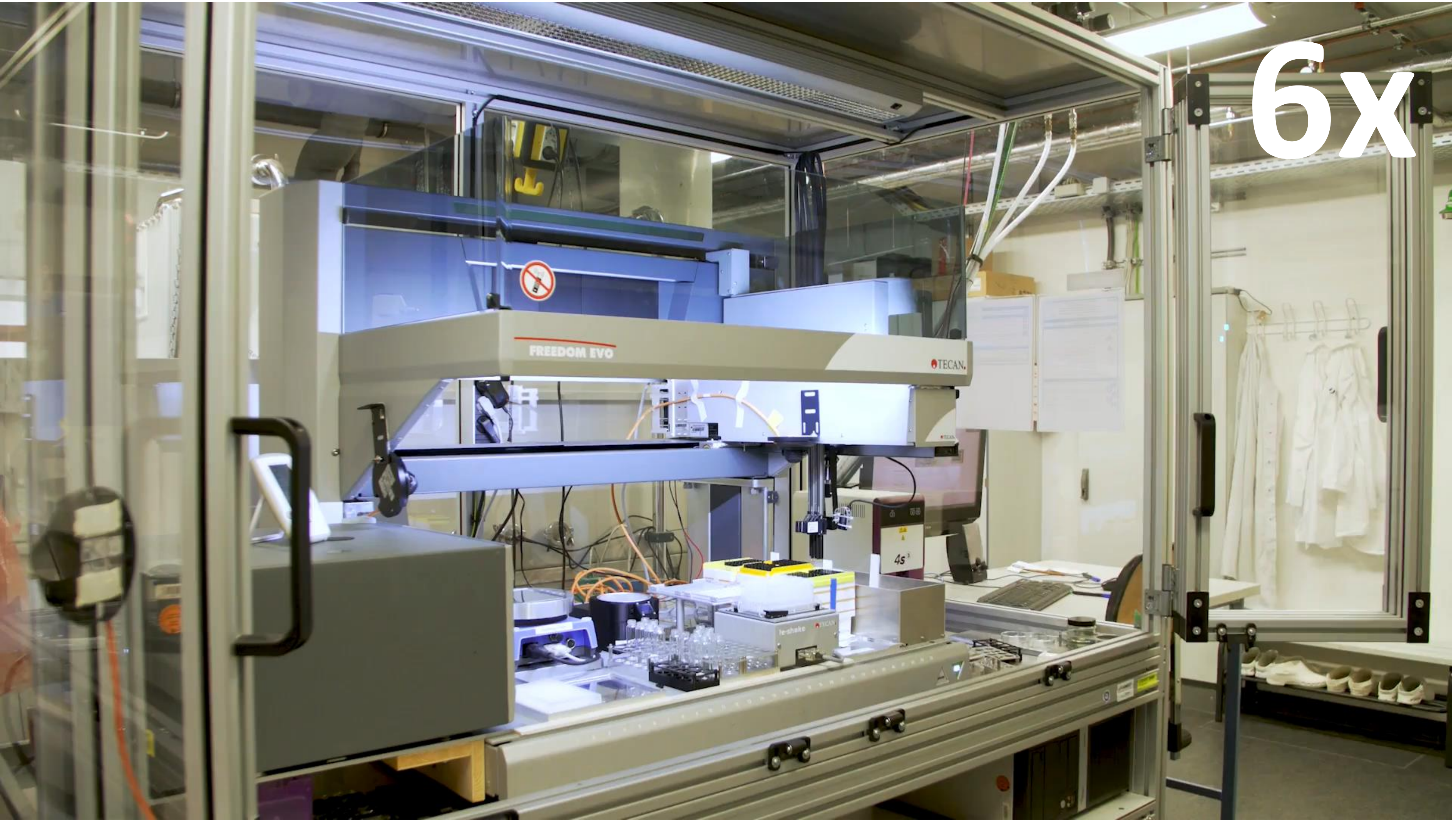

Oleksandra Raievska

Oleksandr Stroyuk

- Ca. 10 samples per day
- Proof of principle
- Photoluminescence as Key Performance Indicator

Cooperation with BAM

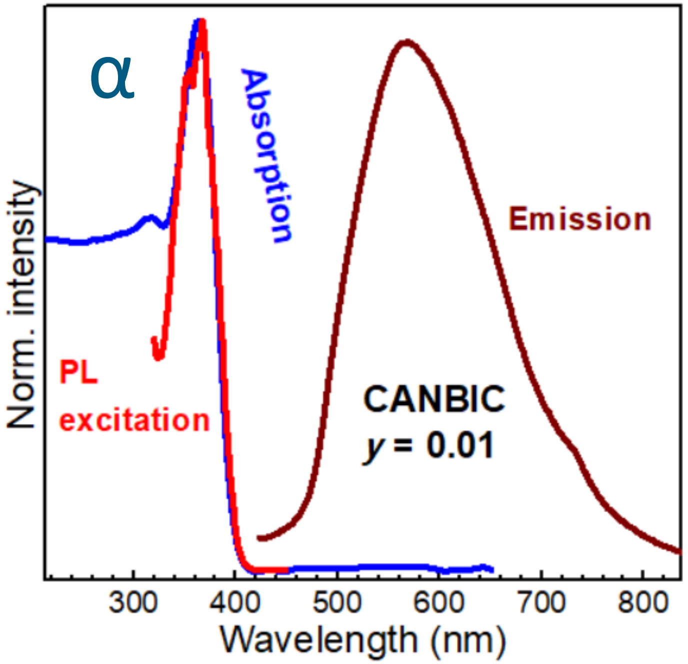
# Automated synthesis of lead free Perovskites



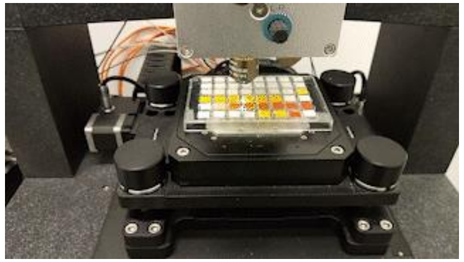

- Ca. 200 samples per day
- Mixing of precursors
- Synthesis
- Drop-casting discrete films
- Analysis by PL
- Manual exchange between stations

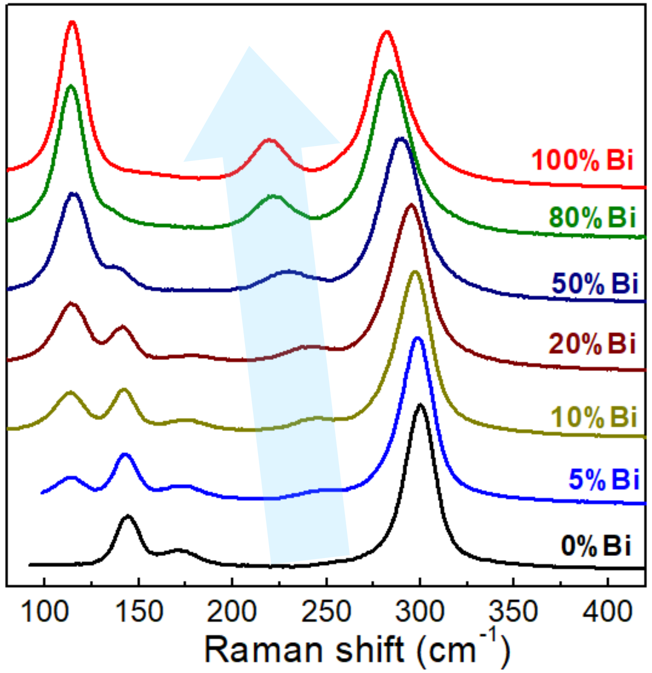
- Exploitation and optimization
- Grid search
- Detailed and systematic follow-up characterization

# High-throughput Characterization $Cs_2(Na,Ag)Bi_yIn_{1-y}Cl_6$

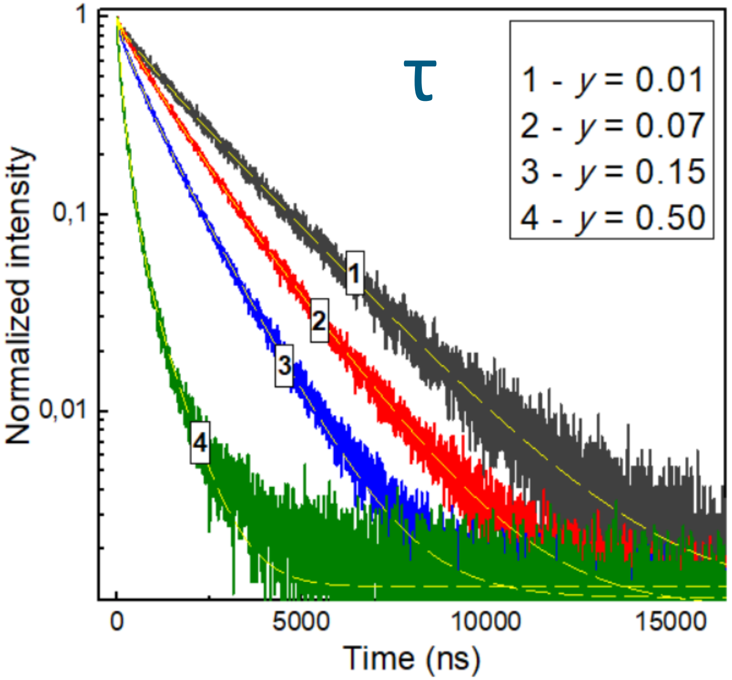



**UV-Vis Absorption**
**Spectral Photoluminescence**



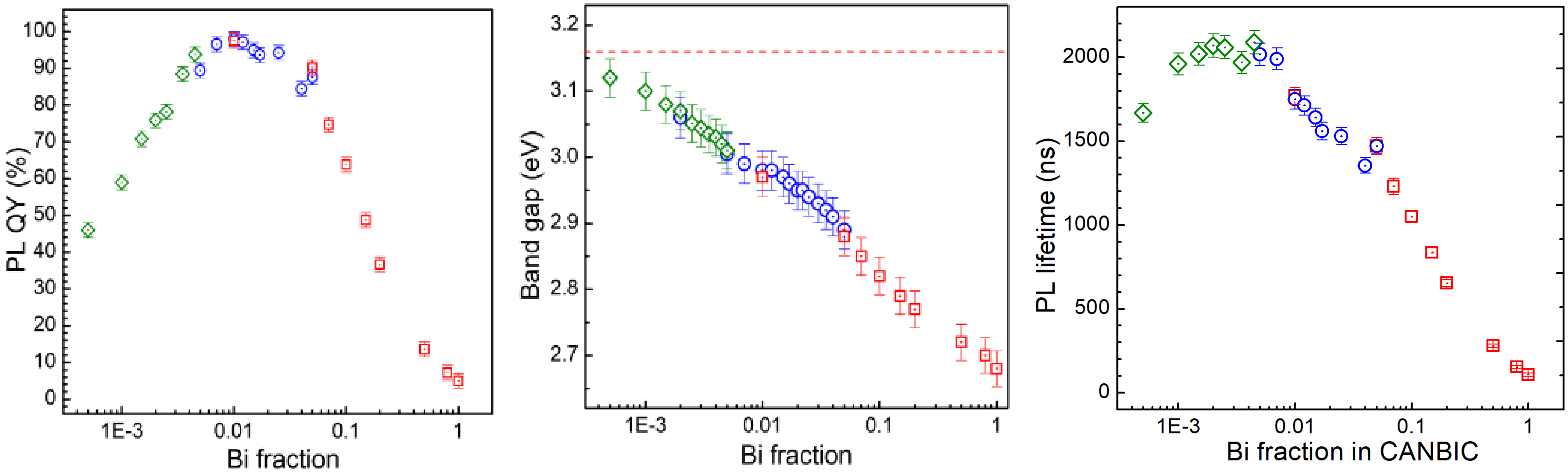



**Raman**




**Time Resolved**
**Photoluminescence**

# Detailed Results $Cs_2(Na,Ag)Bi_yIn_{1-y}Cl_6$



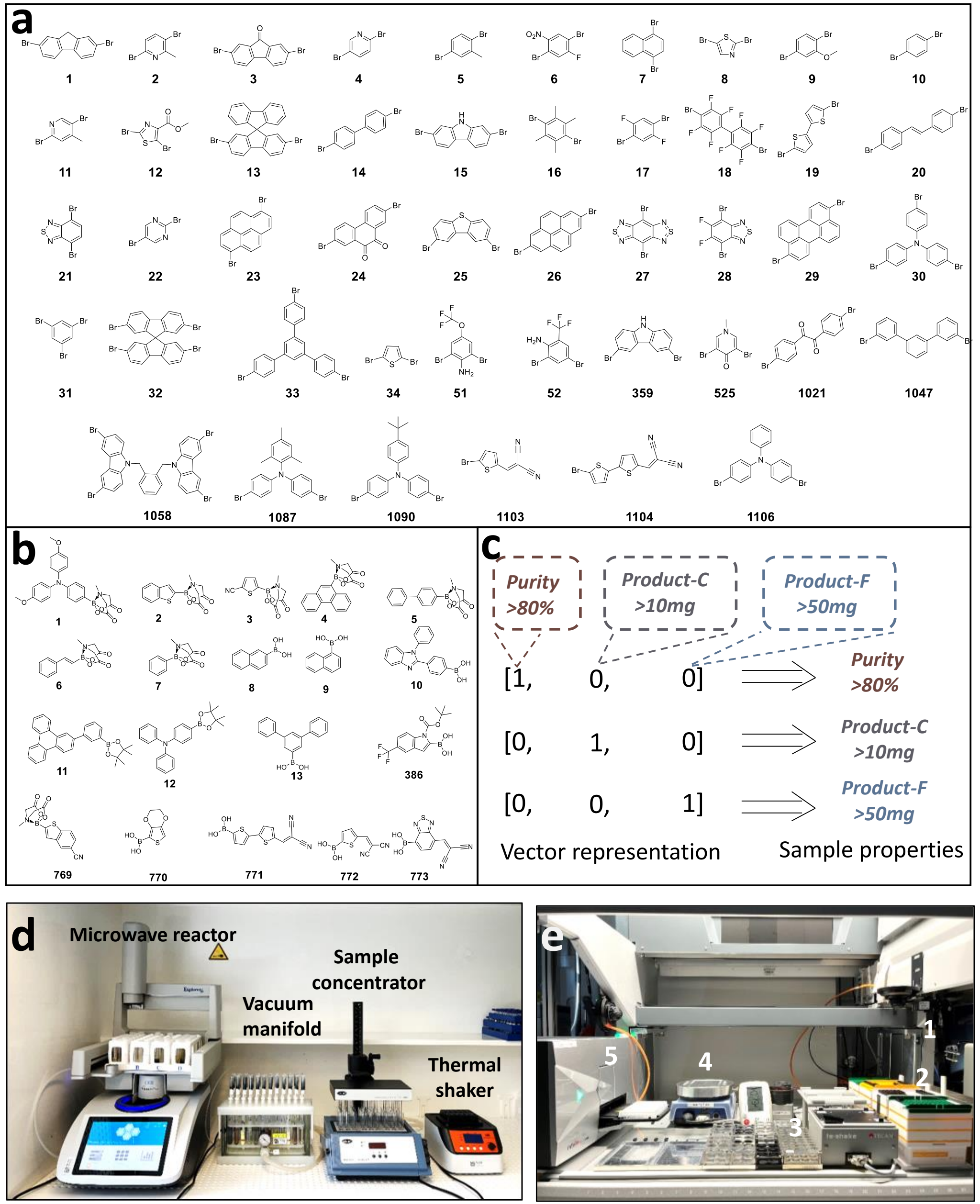

- Variation of Bi-fraction by 4 orders of magnitude
- Outstandingly high photoluminescent quantum yield (close to 100%)
- Tunable Bandgap
- Long charge carrier lifetime (>1µs for more than 2 orders of magnitude in Bi-fraction)
- Very high stability

(1) Stroyuk, O. L. et al., Cs2AgxNa1-xBiyIn1-yCl6 Perovskites Approaching Photoluminescence Quantum Yields of 100 %. Materials Advances 3, 7894-7903 (2022). DOI: 10.1039/D2MA00737A
(2) Stroyuk, O. et al., "Green" synthesis of highly luminescent lead-free Cs2AgxNa1−xBiyIn1−yCl6 perovskites. Journal of Materials Chemistry C 10, 9938-9944 (2022). DOI: 10.1039/D2TC02055F

# Automated synthesis of small molecules

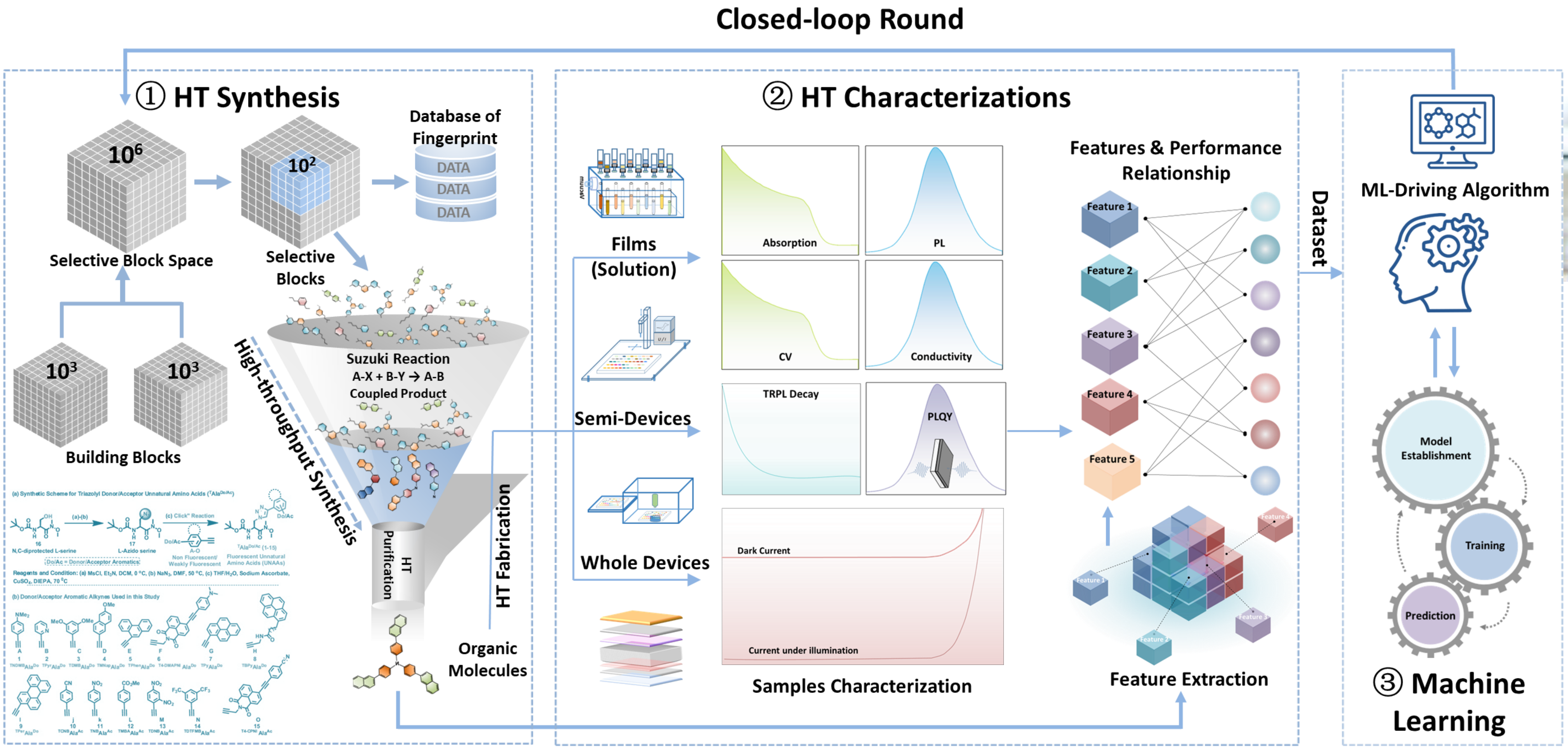
- About 40 samples per week
- Drop-casting discrete films
- Analysis by PL, UV/Vis, EC, DFT, ...
- Extended purity analysis
- Combinatorial approach in the moment
- Guidance by DFT
- Hybrid selection of precursors

**J. Wu et al.**, An integrated system built for small-molecule semiconductors via high-throughput approaches, JACS (2023), doi.org/10.1021/jacs.3c03271




Dr. Jianchang Wu

# From automated synthesis to autonomous optimization of HTL for perovskites

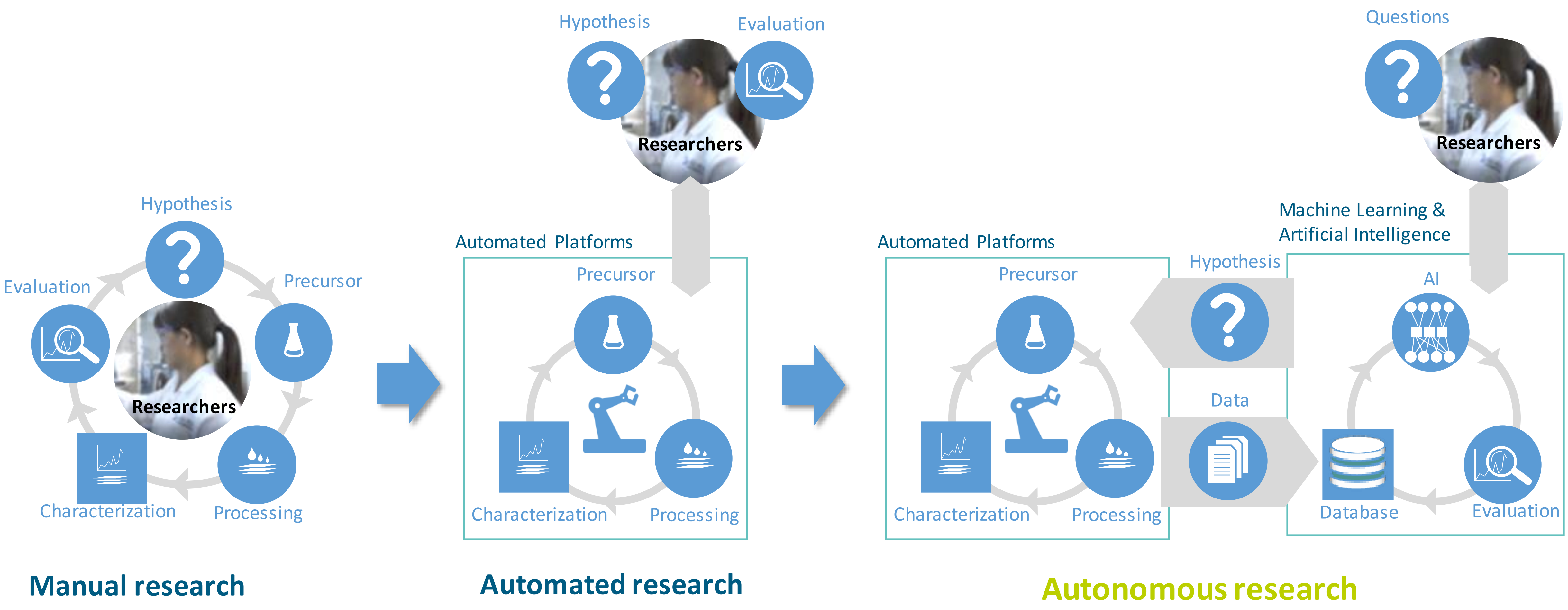



Prof. Pascal Friederich

Karlsruher Institut für Technologie

**J. Wu et al.**, An integrated system built for small-molecule semiconductors via high-throughput approaches, JACS (2023), doi.org/10.1021/jacs.3c03271

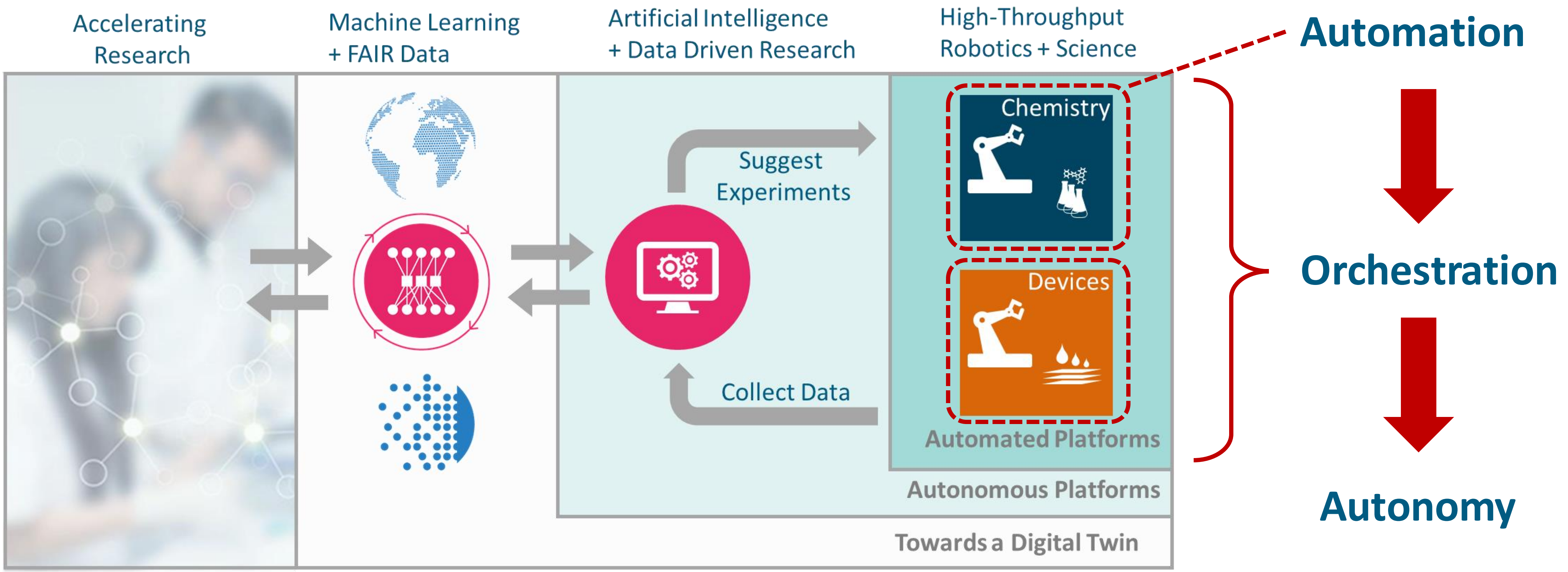
# From Manual to Automated to Autonomous Materials Research: Acceleration




Wagner et al., The evolution of Materials Acceleration Platforms: toward the laboratory of the future with AMANDA, Journal of Materials Science (2021), https://doi.org/10.1007/s10853-021-06281-7

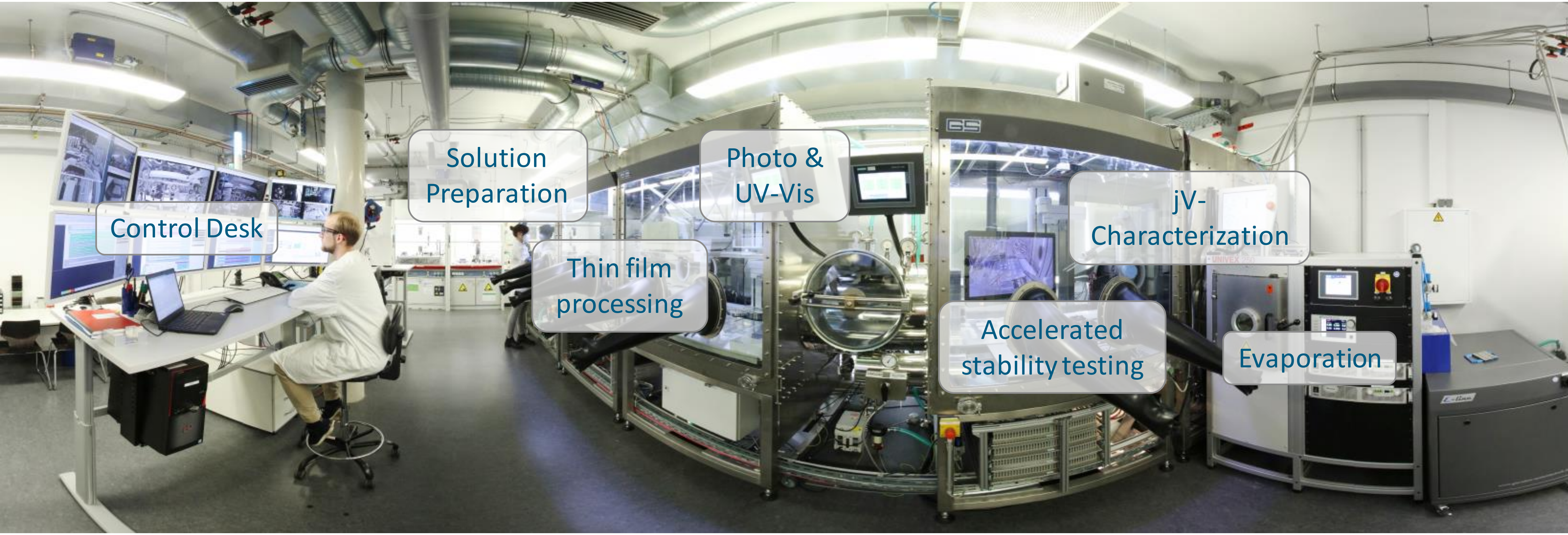
# Autonomous Optimization & Devices

# The path autonomous Materials Discovery




**Automation** – repetitive execution of same task on one piece of equipment
**Orchestration** – execution of tasks on many pieces of equipment in a coordinated workflow
**Autonomy** – intelligent and adaptive workflow planning in order to achieve a target objective

# The platform AMANDA-Line1




- Fully automated handling of substrates, materials and consumables
- Fully integrated manufacturing, characterization and ageing of printed PV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TIP RACK IN
TIP RACK OUT
VIAL HOLDER IN
VIAL HOLDER OUT
Substrate Carrier IN


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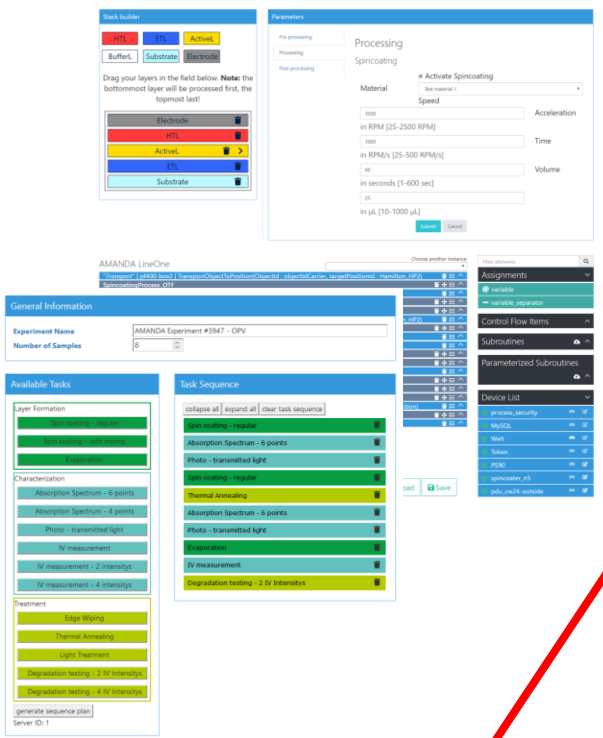
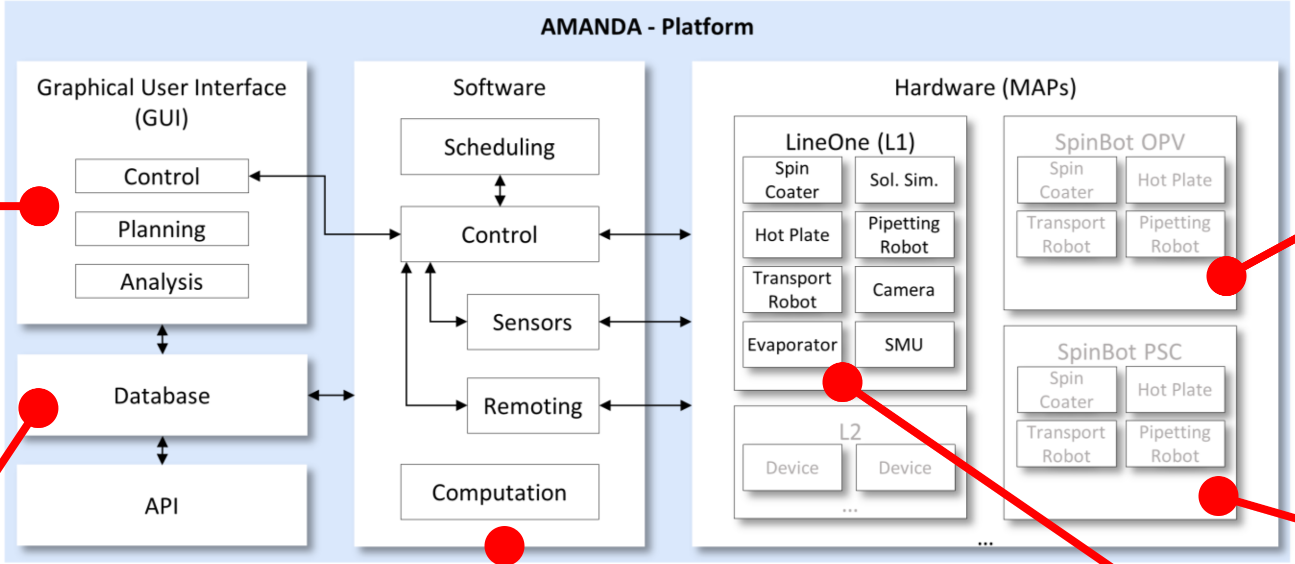
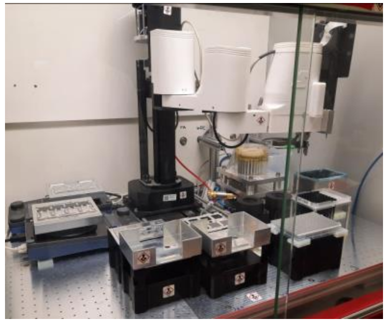
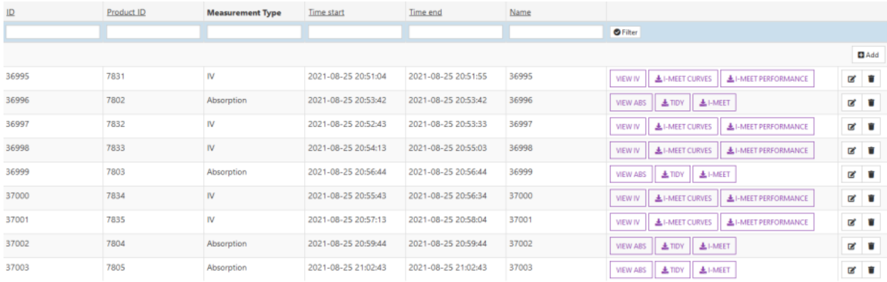
# The AMANDA Platform – multiple MAP control system

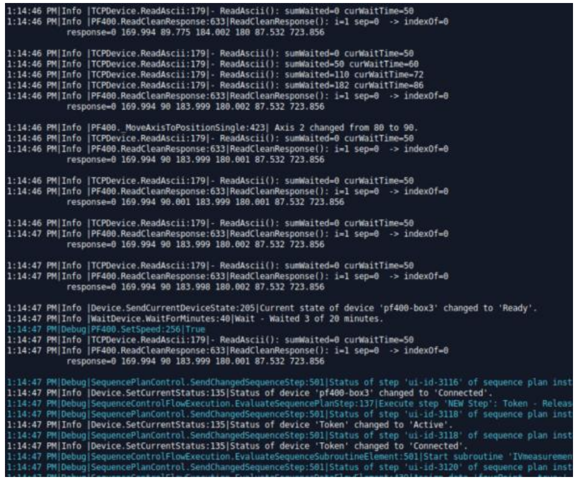






| ID | Product ID | Measurement Type | Time start | Time end | Name | | |
|---|---|---|---|---|---|---|---|
| 36995 | 7831 | IV | 2021-08-25 20:51:04 | 2021-08-25 20:51:55 | 36995 | VIEW IV · I-MEET CURVES · I-MEET PERFORMANCE | |
| 36996 | 7802 | Absorption | 2021-08-25 20:53:42 | 2021-08-25 20:53:42 | 36996 | VIEW ABS · TIDY · I-MEET | |
| 36997 | 7832 | IV | 2021-08-25 20:52:43 | 2021-08-25 20:53:33 | 36997 | VIEW IV · I-MEET CURVES · I-MEET PERFORMANCE | |
| 36998 | 7833 | IV | 2021-08-25 20:54:13 | 2021-08-25 20:55:03 | 36998 | VIEW IV · I-MEET CURVES · I-MEET PERFORMANCE | |
| 36999 | 7803 | Absorption | 2021-08-25 20:56:44 | 2021-08-25 20:56:44 | 36999 | VIEW ABS · TIDY · I-MEET | |
| 37000 | 7834 | IV | 2021-08-25 20:55:43 | 2021-08-25 20:56:34 | 37000 | VIEW IV · I-MEET CURVES · I-MEET PERFORMANCE | |
| 37001 | 7835 | IV | 2021-08-25 20:57:13 | 2021-08-25 20:58:04 | 37001 | VIEW IV · I-MEET CURVES · I-MEET PERFORMANCE | |
| 37002 | 7804 | Absorption | 2021-08-25 20:59:44 | 2021-08-25 20:59:44 | 37002 | VIEW ABS · TIDY · I-MEET | |
| 37003 | 7805 | Absorption | 2021-08-25 21:02:43 | 2021-08-25 21:02:43 | 37003 | VIEW ABS · TIDY · I-MEET | |






Wagner et al., The evolution of Materials Acceleration Platforms: toward the laboratory of the future with AMANDA, Journal of Materials Science (2021), https://doi.org/10.1007/s10853-021-06281-7

# AMANDA GUI – Data compilation

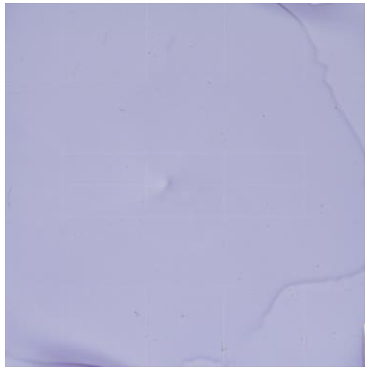
Product Information for Product with ID 7627

**Production Date** 2021-08-12 13:17:08
**Production Size** 2.5x2.5
**Additional Information** created: 8/12/2021 1:17:08 PM

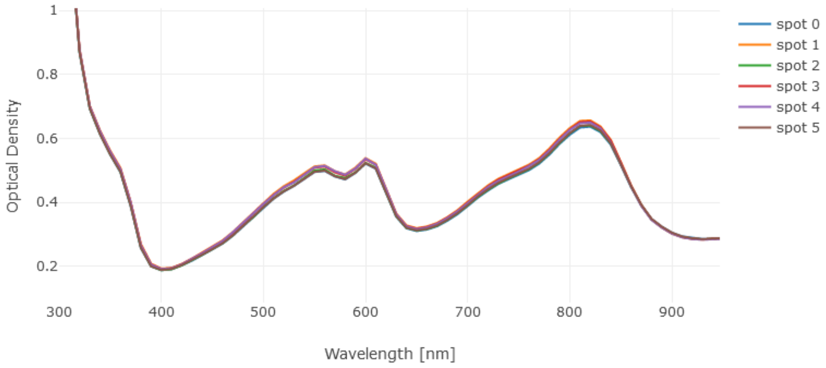
Available Measurements for Selected Product

| ID | Start | End | Type |
|---|---|---|---|
| 33998 | 2021-08-12 13:17:08 | 2021-08-12 13:17:08 | SequencePlan |
| 34010 | 2021-08-12 13:23:49 | 2021-08-12 13:23:49 | SequencePlanEndVariable |
| 34017 | 2021-08-12 13:50:25 | 2021-08-12 13:50:39 | Photo |
| 34023 | 2021-08-12 13:57:16 | 2021-08-12 13:57:16 | Absorption |
| 34048 | 2021-08-12 15:06:35 | 2021-08-12 15:06:35 | SequencePlan |
| 34053 | 2021-08-12 15:20:31 | 2021-08-12 15:35:26 | Evaporation |
| 34054 | 2021-08-12 15:36:27 | 2021-08-12 15:55:05 | Evaporation |
| 34062 | 2021-08-12 16:11:06 | 2021-08-12 16:11:06 | SequencePlan |
| 34093 | 2021-08-12 17:25:28 | 2021-08-12 17:25:28 | SequencePlan |
| 34112 | 2021-08-12 17:57:30 | 2021-08-12 17:58:40 | IV |
| 34133 | 2021-08-12 18:46:55 | 2021-08-12 18:48:06 | IV |
| 34154 | 2021-08-12 19:34:58 | 2021-08-12 19:36:09 | IV |
| 34175 | 2021-08-12 20:51:05 | 2021-08-12 20:52:16 | IV |
| 34196 | 2021-08-12 22:07:12 | 2021-08-12 22:08:22 | IV |

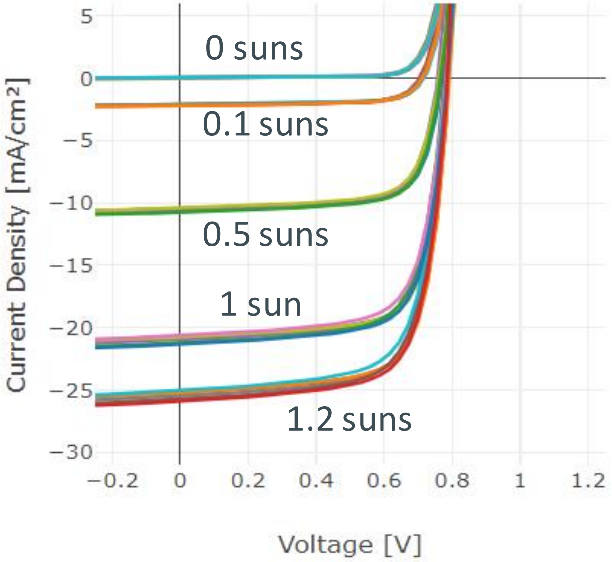
## Image



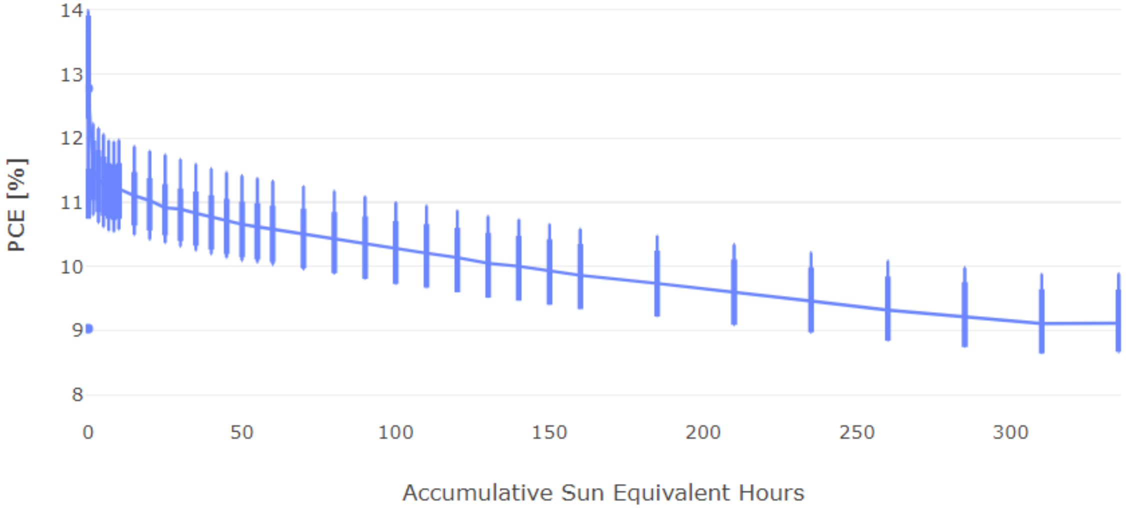
## Absorption Spectrum




## J-V-Curve

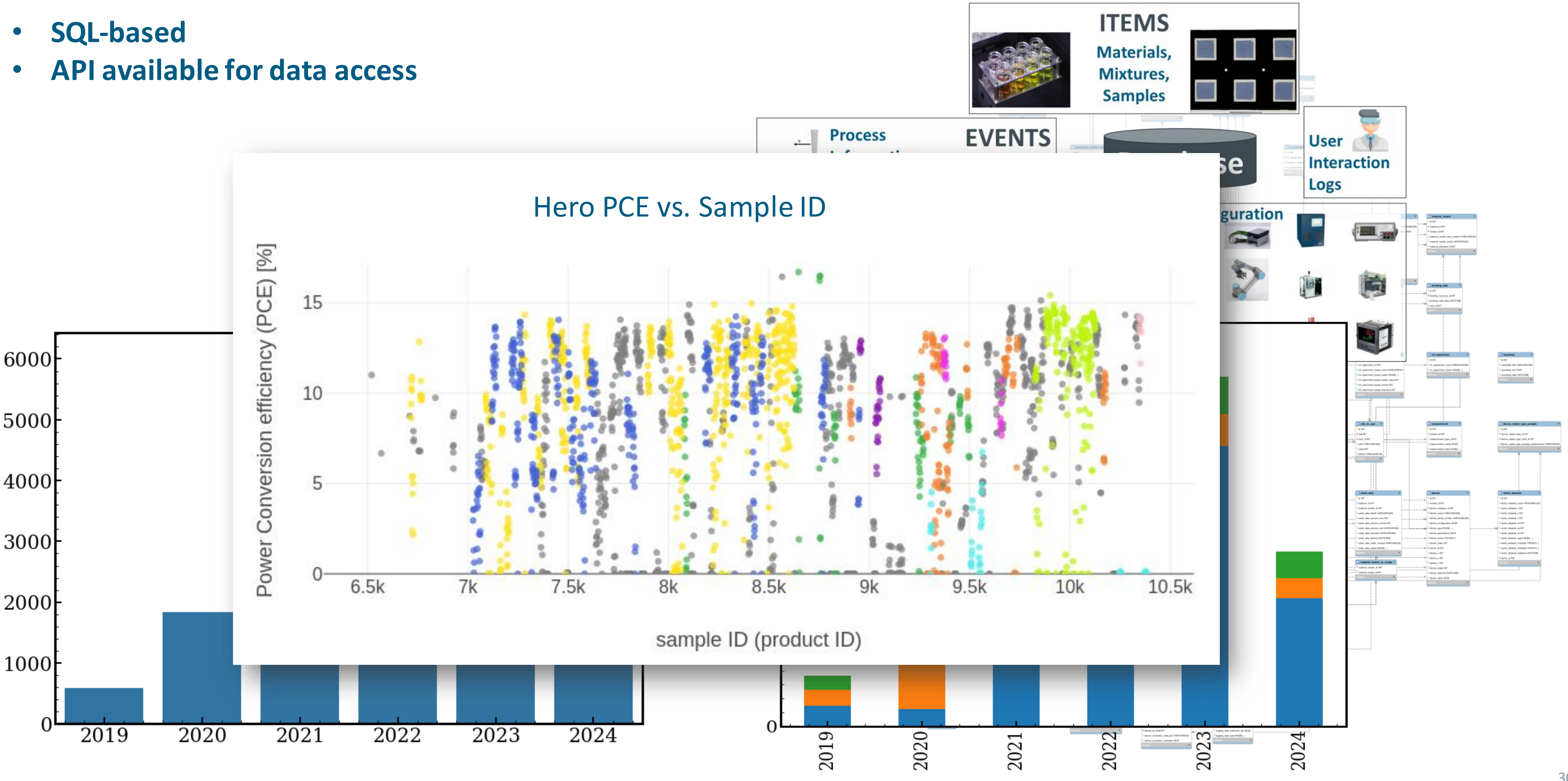



## Stability



# The AMANDA Database

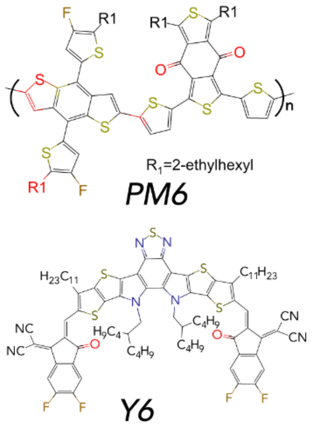
- SQL-based
- API available for data access

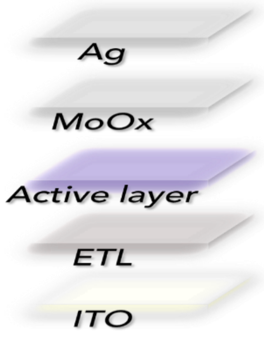


# Automated Cell Manufacturing and Characterization

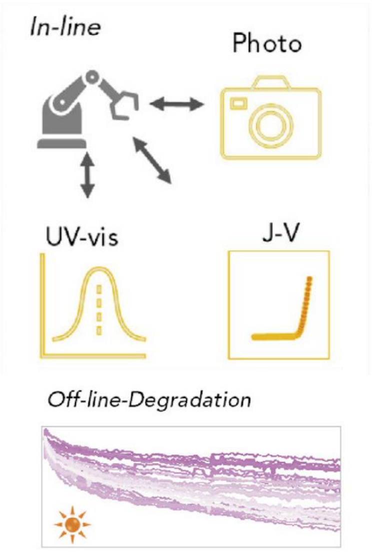
- Robotic-platform for cell manufacturing and characterization
- 116 variations manufactured and characterized
- Absorption spectra of films and jV-Curves of cells measured
- Degradation measured off-line
- Gaussian Process regression used to predict cell performance parameters from absorption spectra after training

## Device Architecture



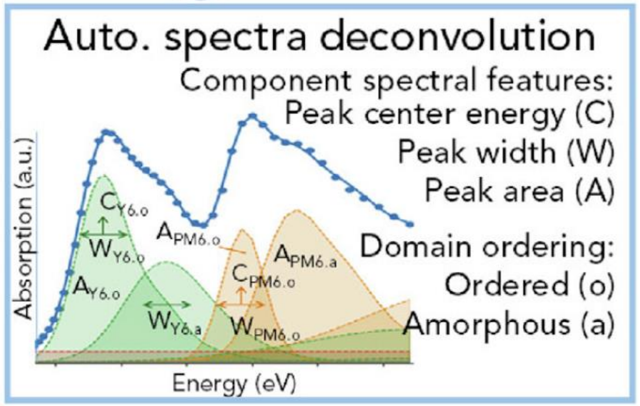




## Characterization




## Parameter Variations

- *DA ratio*
- *Spin-speed*
- *Annealing*
- *Additives*
- *ETL*
- *…*




**Xiaoyan Du**

**Thomas Heumüller**



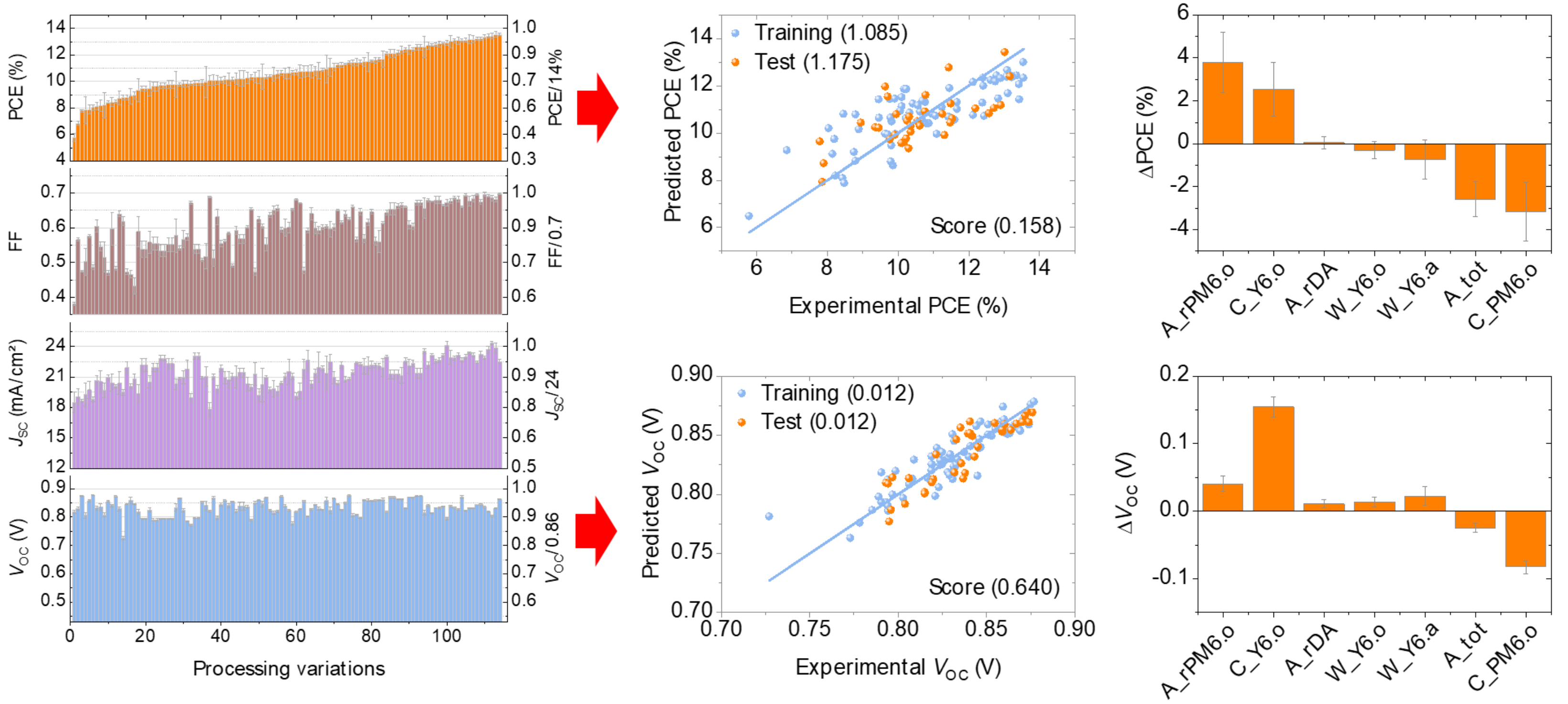
Du et al., Elucidating the Full Potential of OPV Materials Utilizing a High-Throughput Robot-Based Platform and Machine Learning, Joule (2021), https://doi.org/10.1016/j.joule.2020.12.013

# Machine Learning for Cell Optimization

Joule

Performance parameters

Gaussian Process Regression (GPR) prediction




HTCM
Larry Lüer

- 70% of data points used for training, 30% used to test predictive power with GPR
- Very good prediction of Voc with RMSE of 10mV
- Good identification of relevant trends for both Efficiency and Stability
- By now spectral features for 80 materials combinations in our database

Du et al., Elucidating the Full Potential of OPV Materials Utilizing a High-Throughput Robot-Based Platform and Machine Learning, Joule (2021), https://doi.org/10.1016/j.joule.2020.12.013

# Line 1 – Autonomous Operation

## Workflow on AMANDA LINE 1

Spectra

Device Performance

Process Parameters

P1 P2 P3 P4

Bayesian Optimization

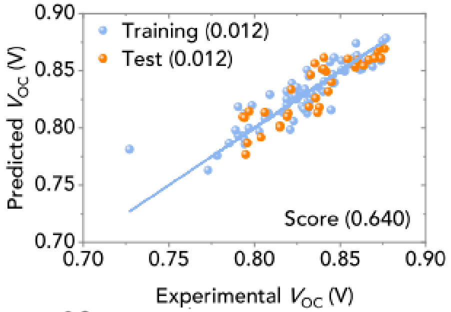
local

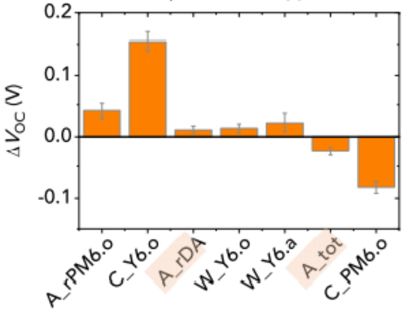
global optimization

## Gaussian Process Regression

All previous experiments

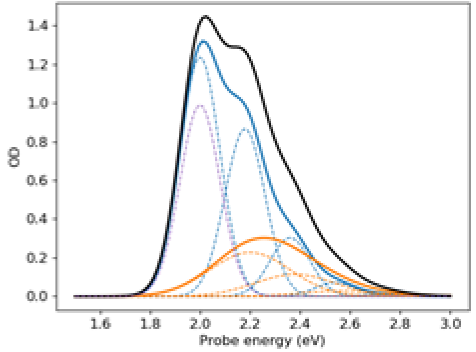
GPR




## Photophysical Modeling

Spectral Modeling




Order

Torsional motion

Domain size

+ Structural motifs

- Train **proxy** to give **nanostructure** from inline experiment
- Predict device performance **without device processing**
- Learn **general design principles**

**Accelerate Bayesian Optimization**

**Find hidden patterns → innovation**

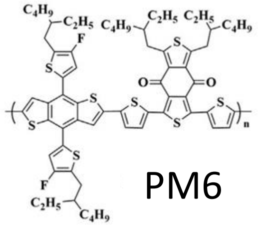

**X. Du** et al., *"Elucidating the Full Potential of OPV Materials Utilizing a High-Throughput Robot-Based Platform and Machine Learning"* , **Joule,** (2021)

**J. Wagner** et al., *"The evolution of Materials Acceleration Platforms: toward the laboratory of the future with AMANDA"* , **J. Mat. Science,** (2021)

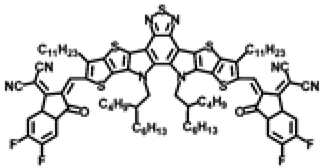
**C. Liu** et al., "Understanding causalities in organic photovoltaics device degradation in a machine learning driven high-throughput platform" , **Adv. Materials,** (2023)

# Line 1 – Autonomous Operation

Optimization of ternary system with 4 variables **> 1000 experiments in Edisonian approach**



PM6

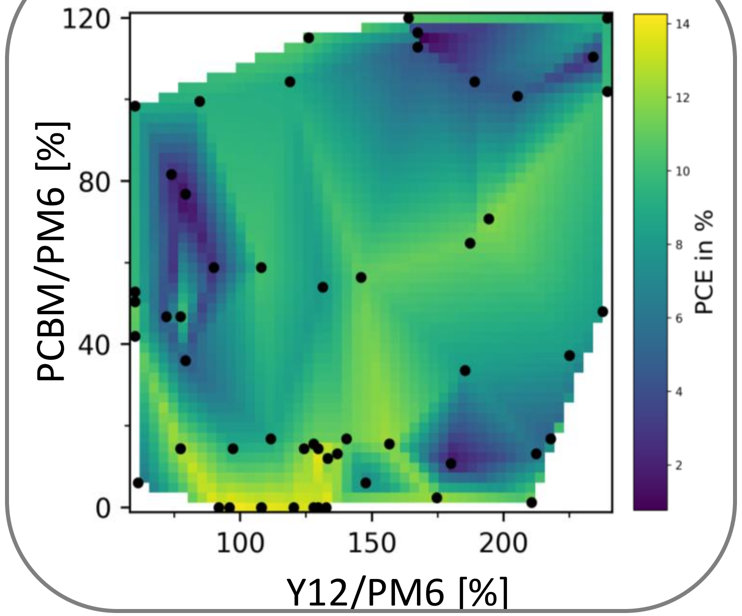


Y12

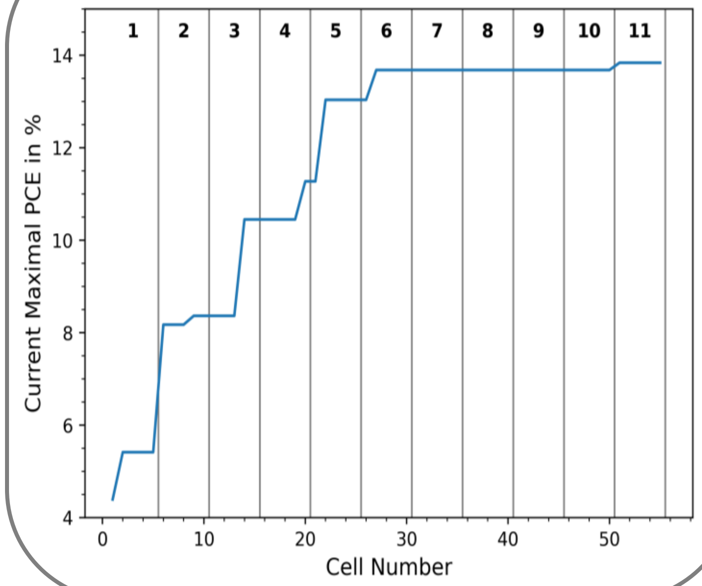
Ternary Additive **PC70BM**

Variables
- Y12 Ratio
- Additive Ratio
- Solvent Ratio (≈ Concentration)
- Spin Speed

Line 1 in autonomous operation **found two optima**, one binary and one ternary




Line 1 in **autonomous operation needed 30 experiments** to find optimium




**Acceleration by a factor of more than 30!**

**T. Osterrieder** et al., *"Artificial intelligence guided organic Solar Cell Device Optimization of a ternary active layer"*, **Energy & Environmental Science,** (2023)

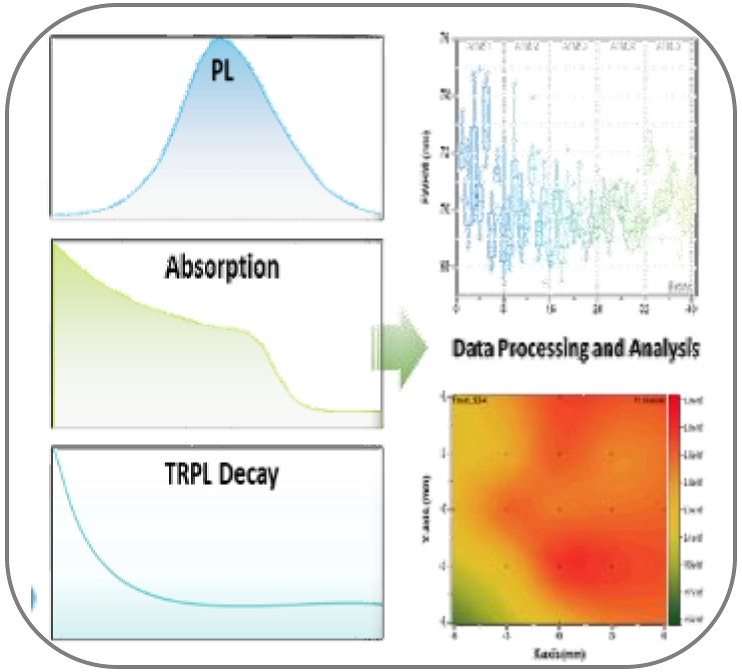
**F. Schmitt** et al., *"Using Machine Learning to optimize the Efficiency and Stability of Organic Solar Cells"*, **MSc Thesis,** (2022)

**S. Langner** et al., *"Beyond ternary OPV: high-throughput experimentation and self-driving laboratories optimize multicomponent systems"*, **Advanced Materials ,** (2020)

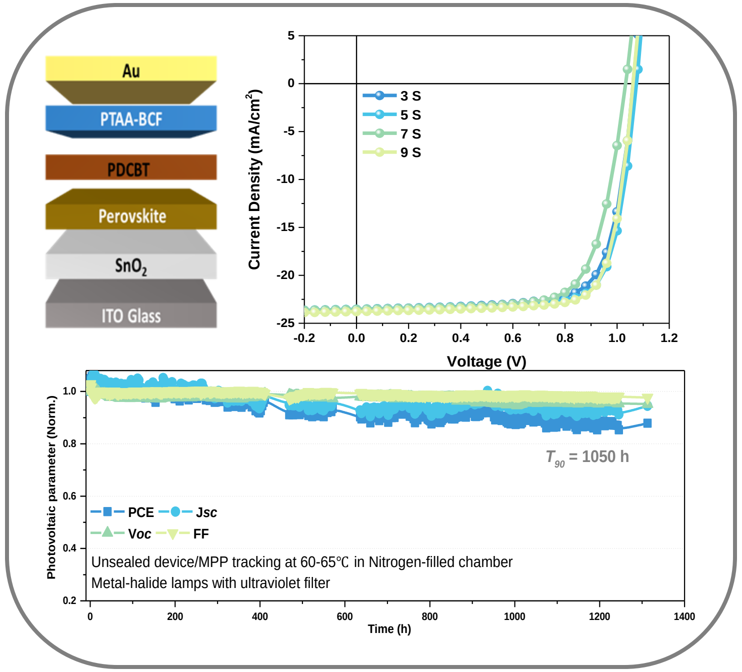
# Line 2 – Autonomous Perovskite Optimization

Controlling environmental processing of perovskites by optimizing **70 process parameters**


Optimization was **purely driven by optical measurements** – dominantly PL (tr, ss, imaging)




Optical optimization resulted in larger area perovskite devices with 20 % and ISOS L T80 (65 C) > 2000 h




- **Acceleration by a factor of more than 50!**

**HIERN**, *"SCIPRIOS licenses SpinBot Technology from FZJ/HIERN platform"*, (2021)

**J. Zhang** et al., *"Exploring the steric hindrance ... In of Quasi-2D perovskites in a high throughput platform"*, **Advanced Functional Materials,** (2022)

**J. Zhang** et al., *"A Fully Robotic Platform for Optimizing the High-dimensional Processing Parameter Space of Perovskite Thin-film Devices"*, , **Advanced Functional Materials,** (2024)

# Technology Acceleration

Solar TAP

# ACCELERATE TRANSFER OF EMERGING-PV TECHNOLOGIES

SOLARTAP Technology. Transfer.

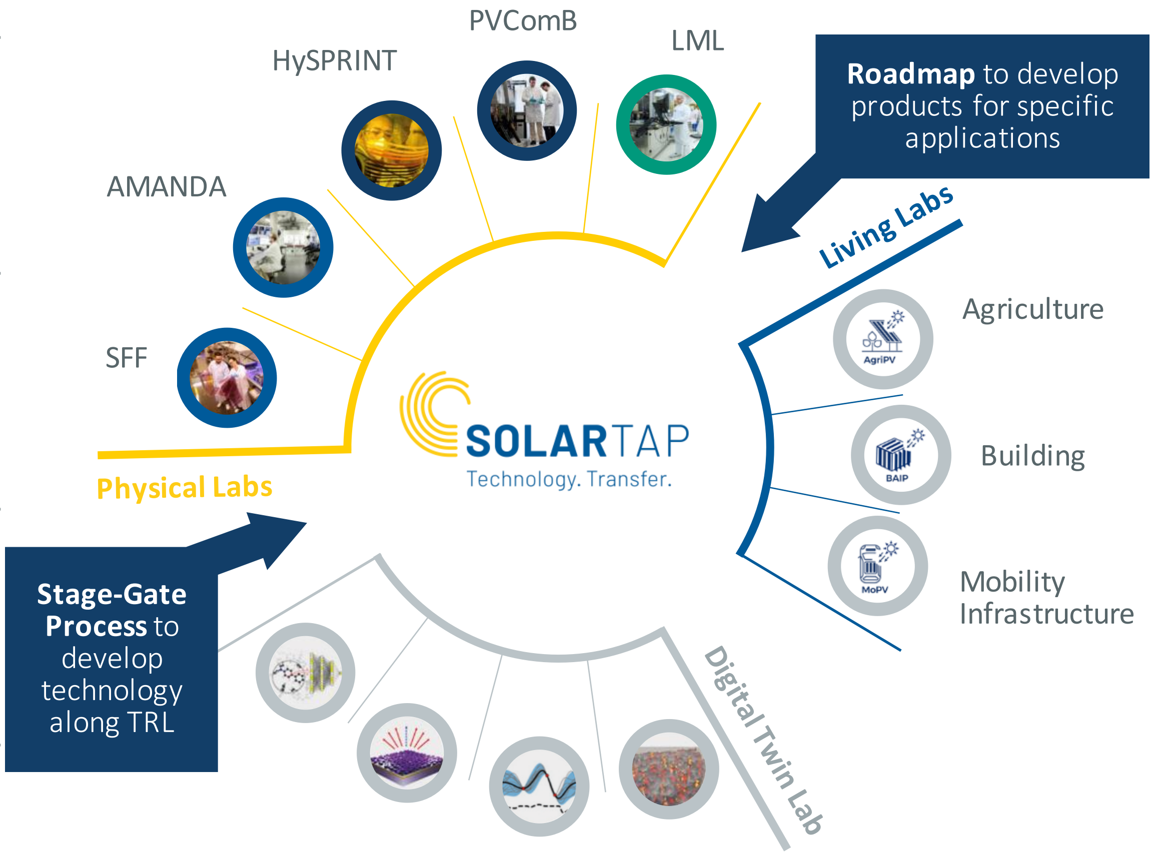
*From Materials Acceleration to Technology Acceleration*

JÜLICH Forschungszentrum

HZB Helmholtz Zentrum Berlin

KIT Karlsruhe Institute of Technology

Helmholtz Scientific Environments




Solar TAP Industry Network

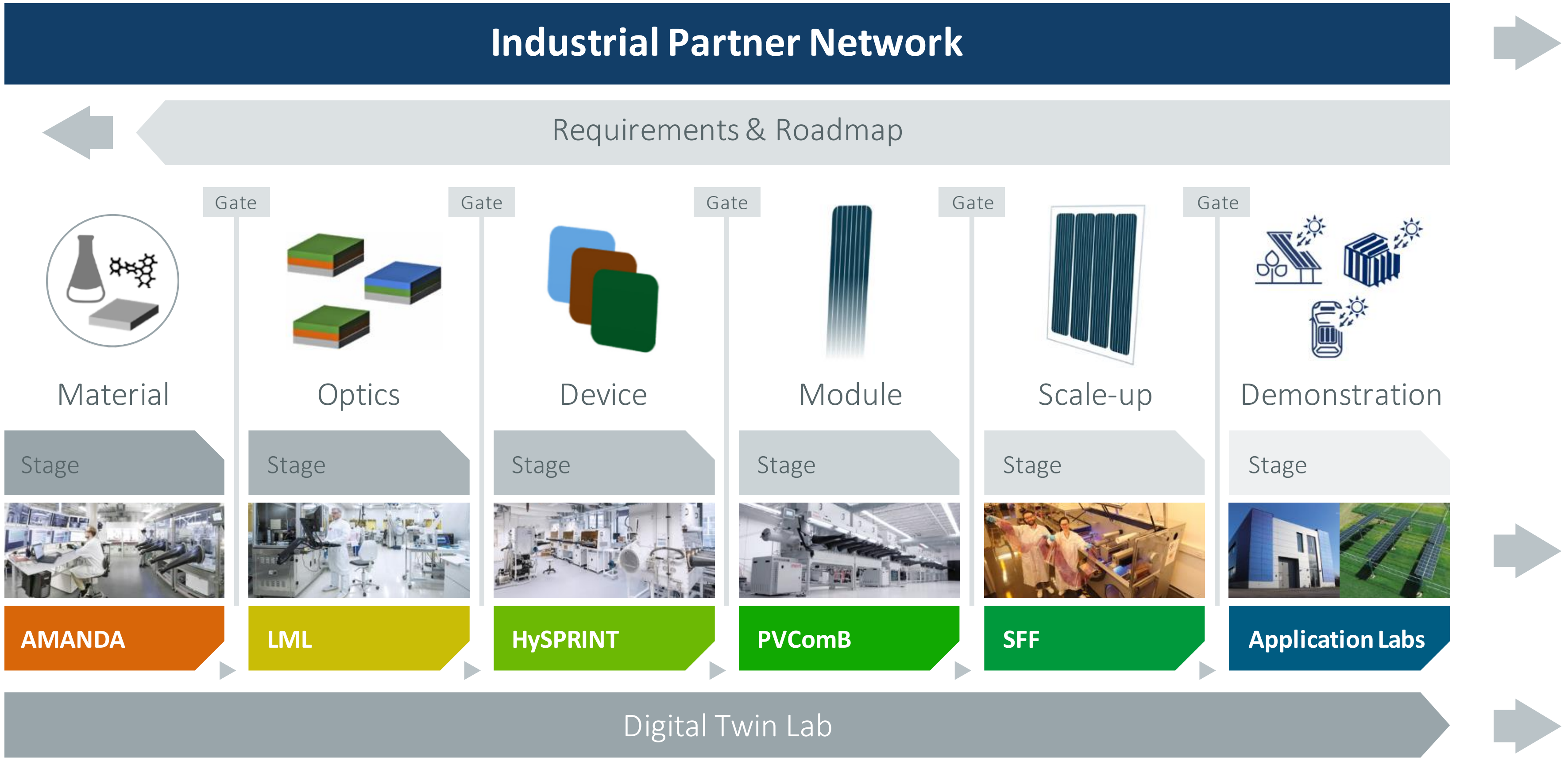
Multi-Benefit Photovoltaic Industry

Solar TAP

# BIDIRECTIONAL APPROACH TO ENABLE MULTI-BENEFIT PV

SOLARTAP Technology. Transfer.

*Transferring technologies along the full value chain.*




Multi-Benefit
Photovoltaic Industry

JÜLICH Forschungszentrum
HZB Helmholtz Zentrum Berlin
KIT Karlsruhe Institute of Technology

# Takeaways

- **Automation** can improve **precision and reproducibility** in materials science
- **Machine learning** can be used to guide the **search** of **complex parameter spaces** for optimal performance of functional materials
- We have developed a **comprehensive database** allowing us to perform **data driven optimization**
- We can **address and solve relevant problems** in materials science and photovoltaics
- Our approach of combining Automation, ML and PV-knowhow **accelerates** materials discovery
- Accelerating a technology requires the whole value chain and a market!

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# Thank you for your attention!

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