AI-enhanced Intensity-Modulated Two-Photon excited fluorescence Microscopy (IM2PM)

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Acknowlegements
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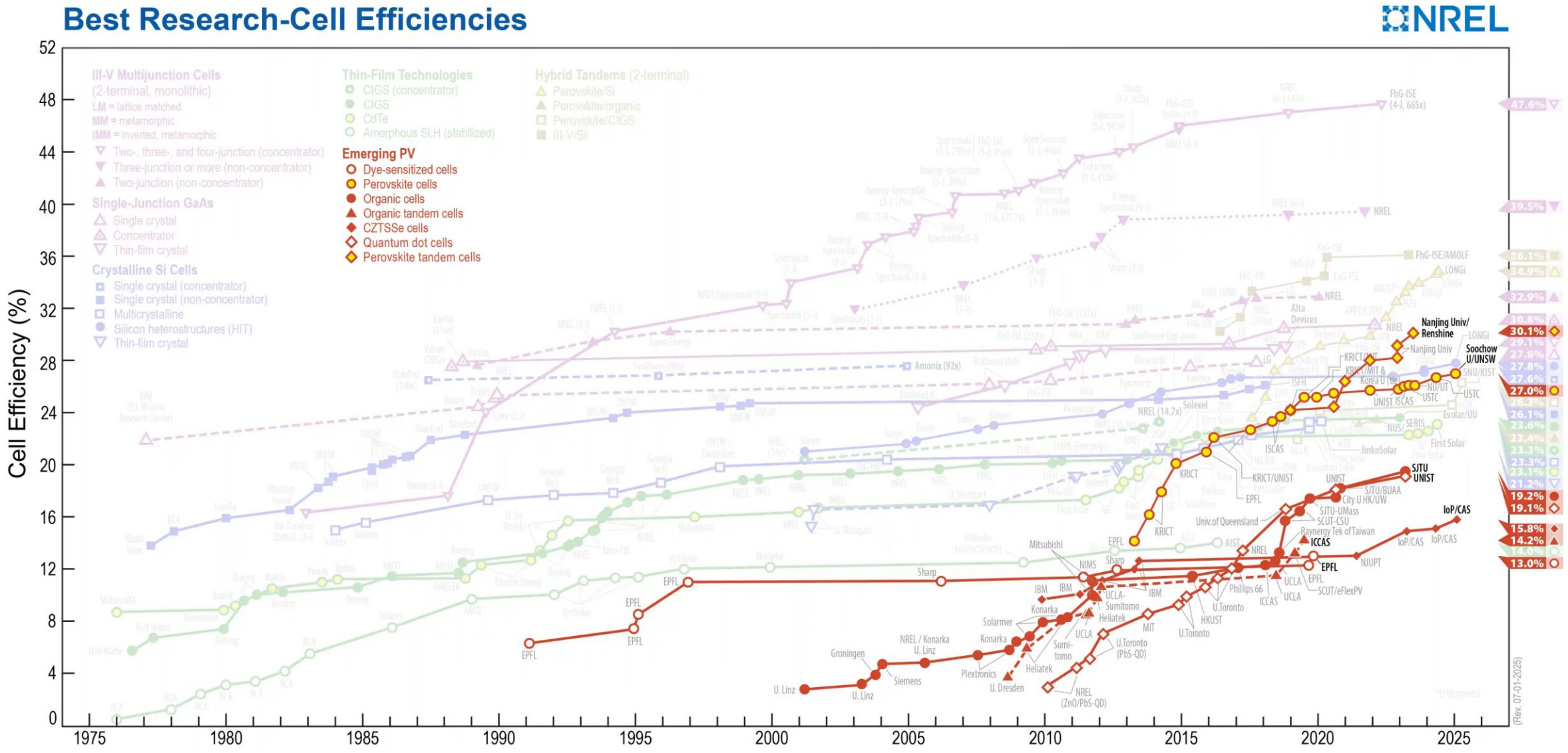
Outlines

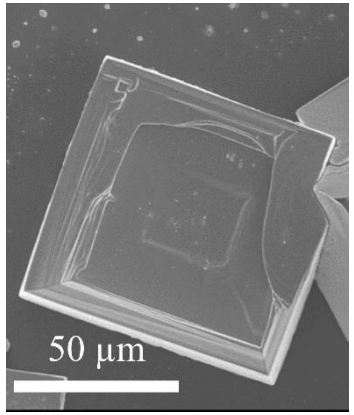
- **Introduction**
- **CCR model** — Charge carrier recombination in perovskite
- **IM2PM** — Nonlinear optical microscopy
- **ml-IM2PM** — Supervised machine learning regression model — pixel vector
- **dl-IM2PM** — (Ongoing) Supervised deep learning regression model — image
- **Conclusion**



1 https://www.nrel.gov/pv/cell-efficiency 2025

Challenges in Perovskite Materials Study

- Defects formation unavoidably
- Morphology heterogeneity
- Understanding of defect formation and the performance of the materials



SEM image of $MAPbBr_3$ perovskite

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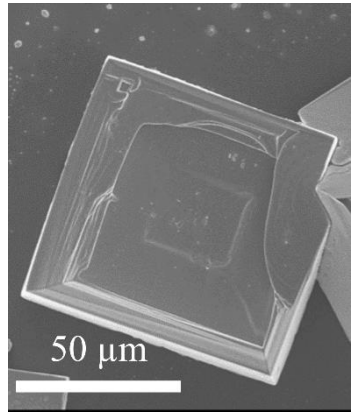
Intensity Modulation Two-Photon Photoluminescence Microscopy (IM2PM)[1]

- Time-saving
- High-resolution mapping (μm scale)
- Photo-induced charge carrier trapping and dynamics information revealed by IM2PM



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1 Shi, Q., Kumar, P. and Pullerits, T., 2023. ACS Physical Chemistry Au, 3(5), pp.467-476.

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- Time-saving
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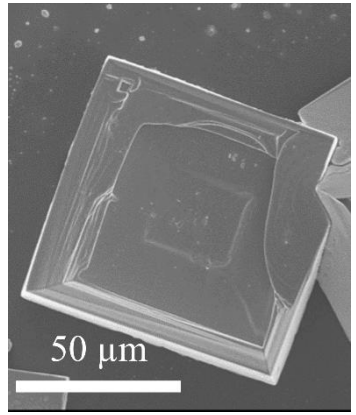
AI-Enhanced IM2PM[2,3]

- Data-driven simulation based on a physical model
- Bridging the photo-induced charge carrier trapping and dynamics with complex experimental data



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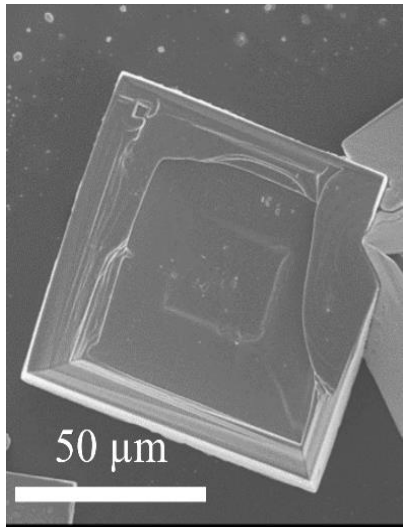
2 Shi, Q., and Pullerits, T., . *ACS Photonics* 11, no. 3 (2024): 1093-1102.
3 Shi, Q., & Pullerits, T. (2025). Energy & Environmental Materials, e70062.



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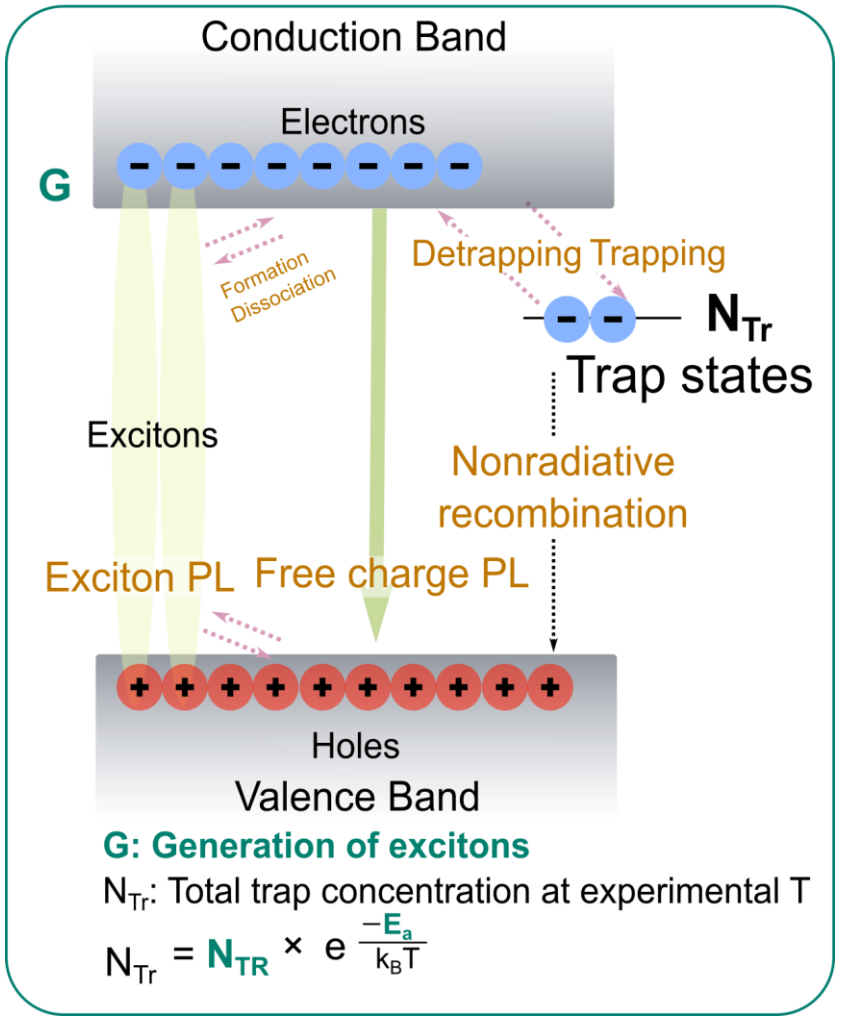
Model parameters

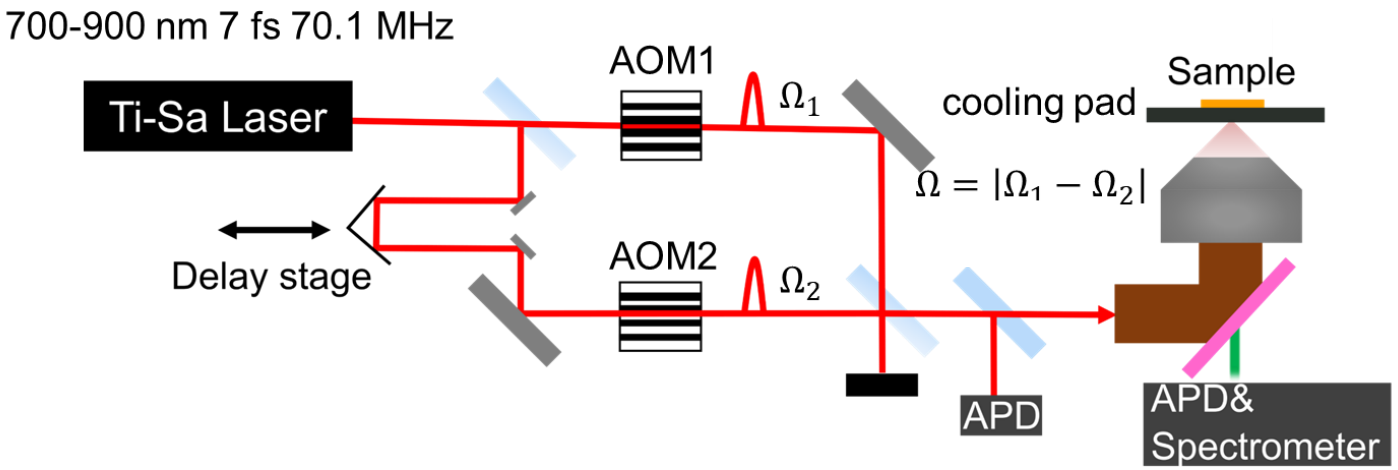


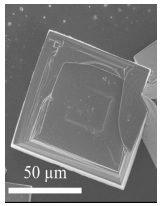
G: Generation of excitons

N_{Tr}: Total trap concentration at experimental T

$$N_{Tr} = N_{TR} \times e^{\frac{-E_a}{k_B T}}$$

Balanced Mach–Zehnder interferometer





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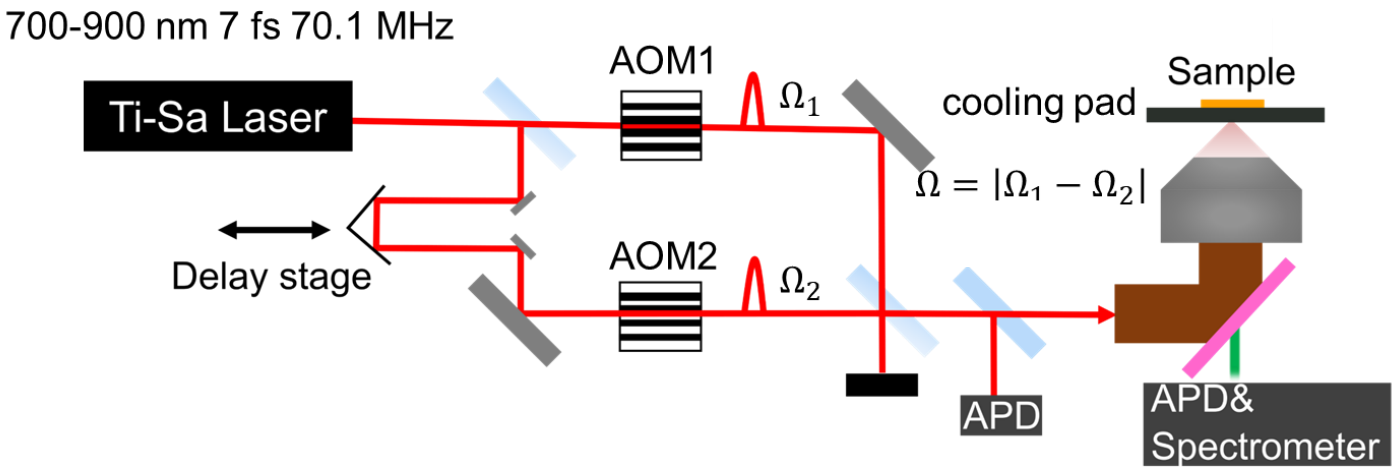

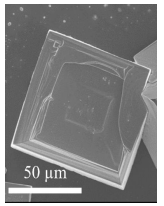

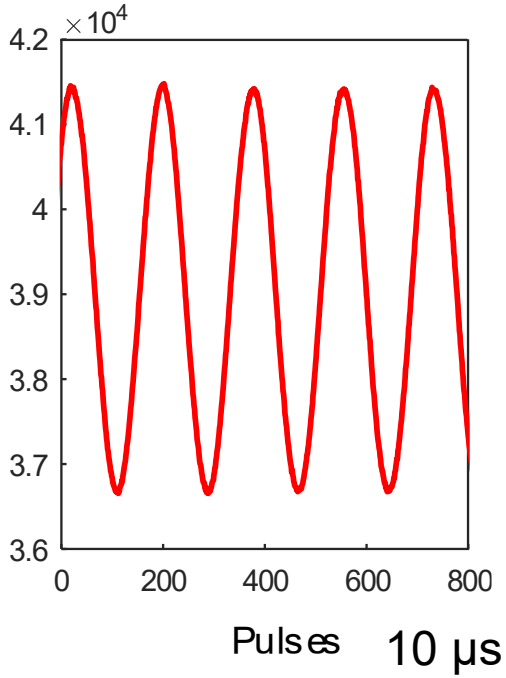

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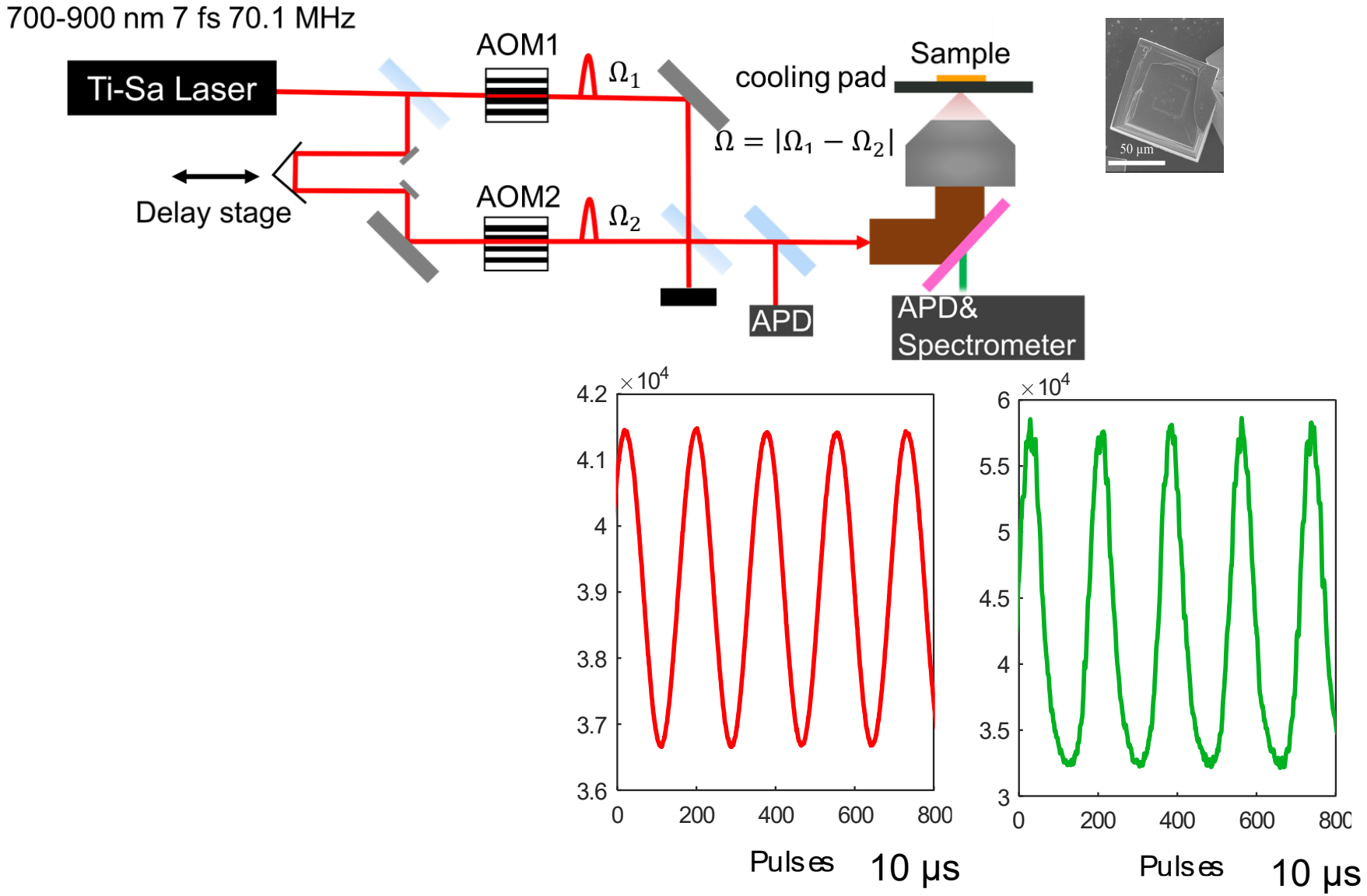


Dataset Simulation

Selection of model parameters (G, E_a, N_{TR})

Y

Parameters sampled

IM2PM Simulation

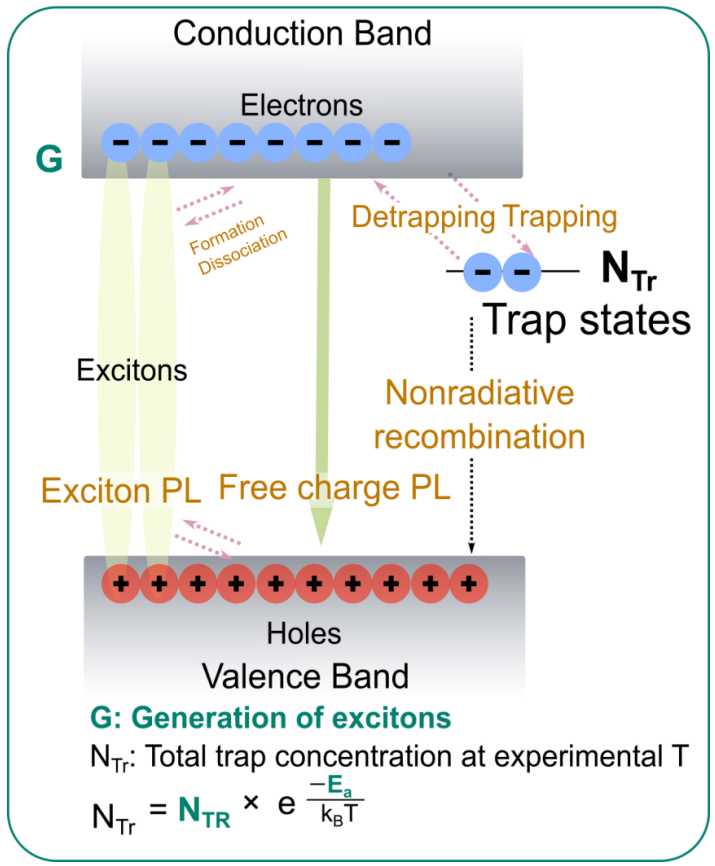
Physics

ODEs+Intensity Modulation

X

Simulated PL (AiH) spectra

Model parameters



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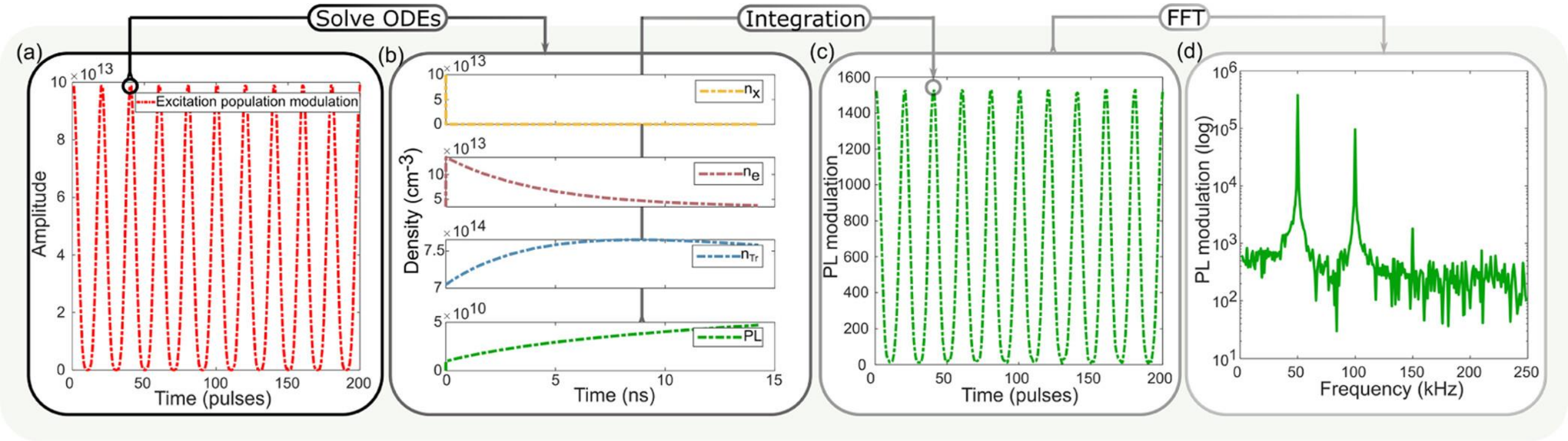

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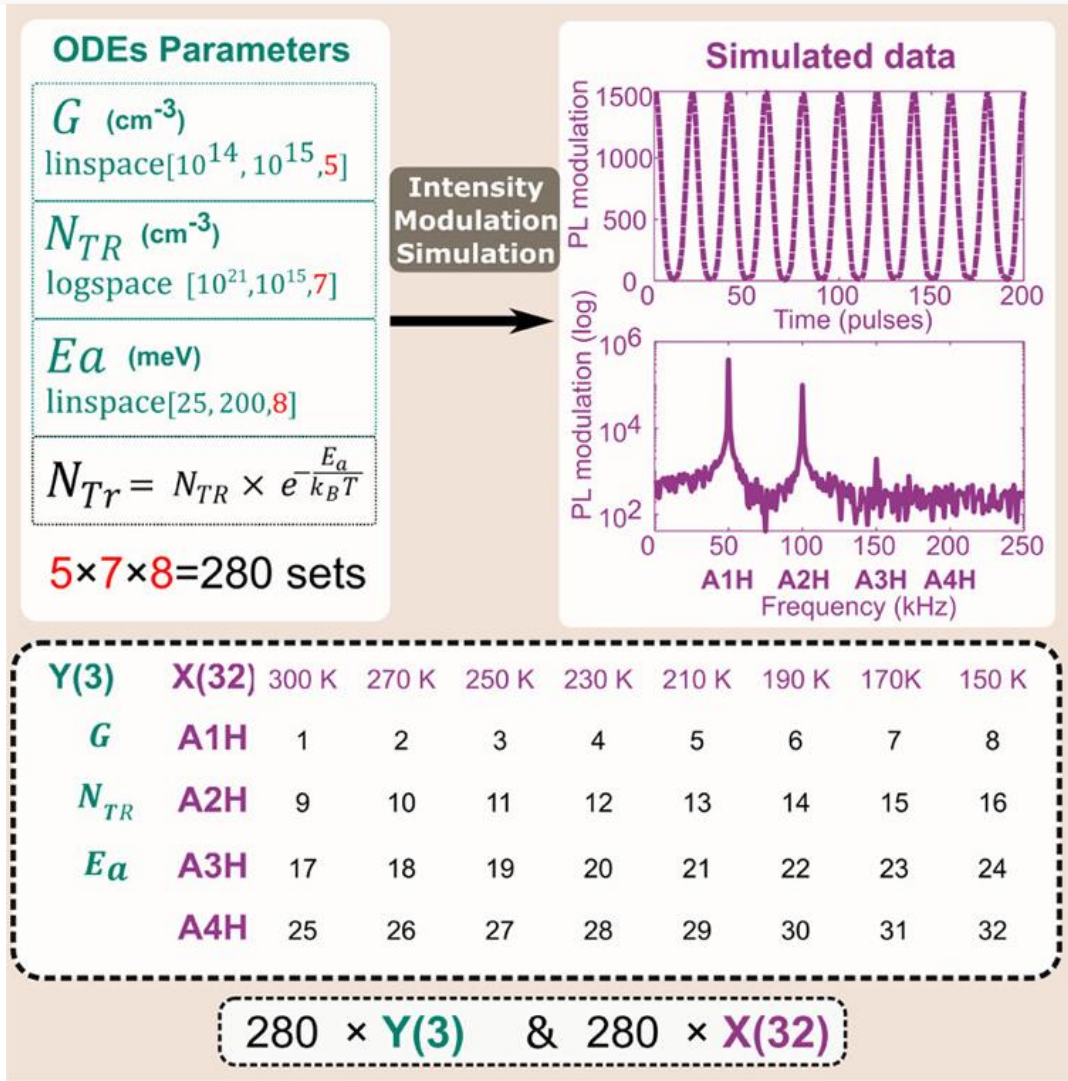
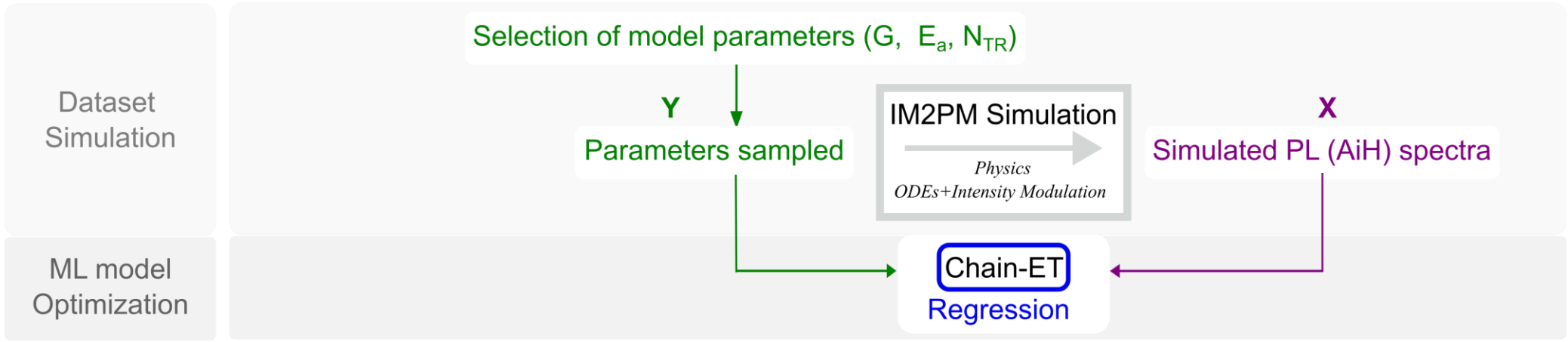


Y(3)	X(32)	300 K	270 K	250 K	230 K	210 K	190 K	170K	150 K
G	A1H	1	2	3	4	5	6	7	8
N_{TR}	A2H	9	10	11	12	13	14	15	16
Ea	A3H	17	18	19	20	21	22	23	24
	A4H	25	26	27	28	29	30	31	32

280 × Y(3) & 280 × X(32)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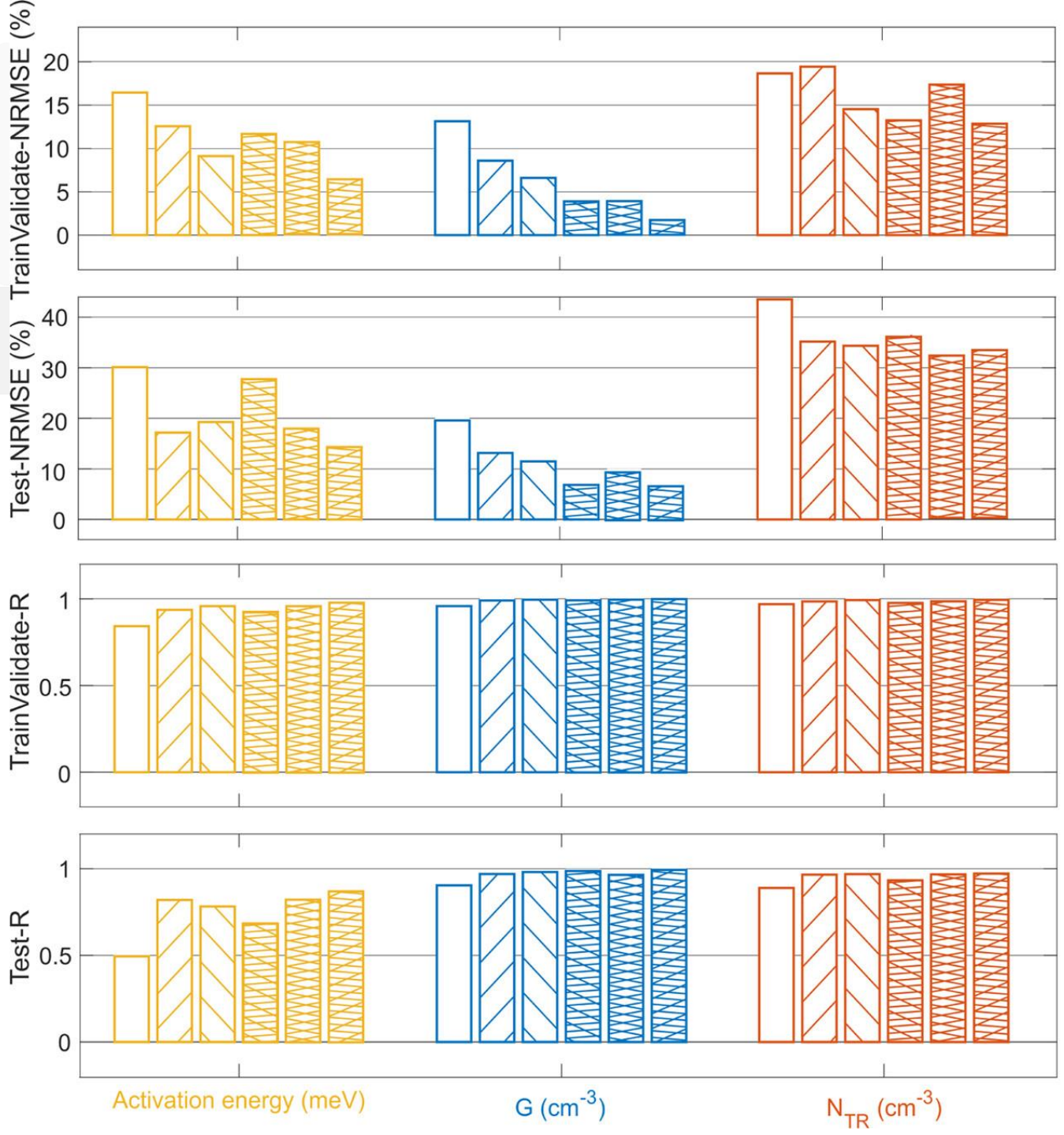
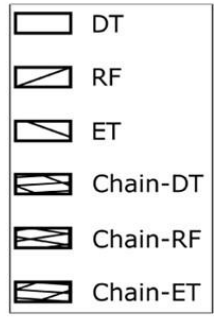
ML model Optimization
Chain-ET
Regression

Dataset Simulation

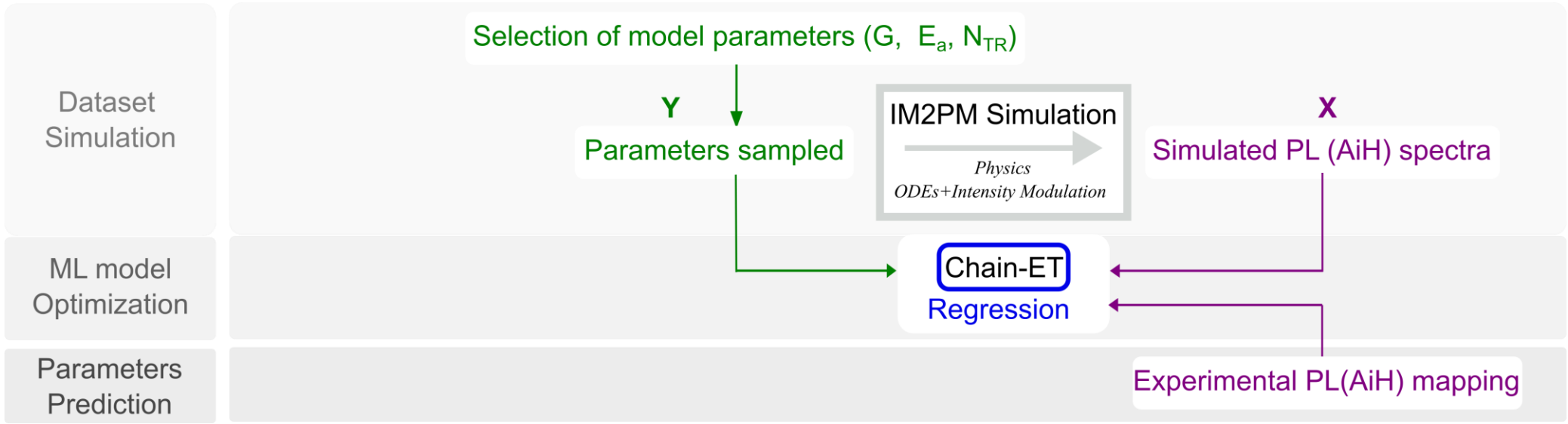
ML model Optimization

ctra





DT: Decision tree
RF: Random forest
ET: Extra tree
Chain: Regression Chain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Experimental PL(AiH) mapping

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ML model Optimization

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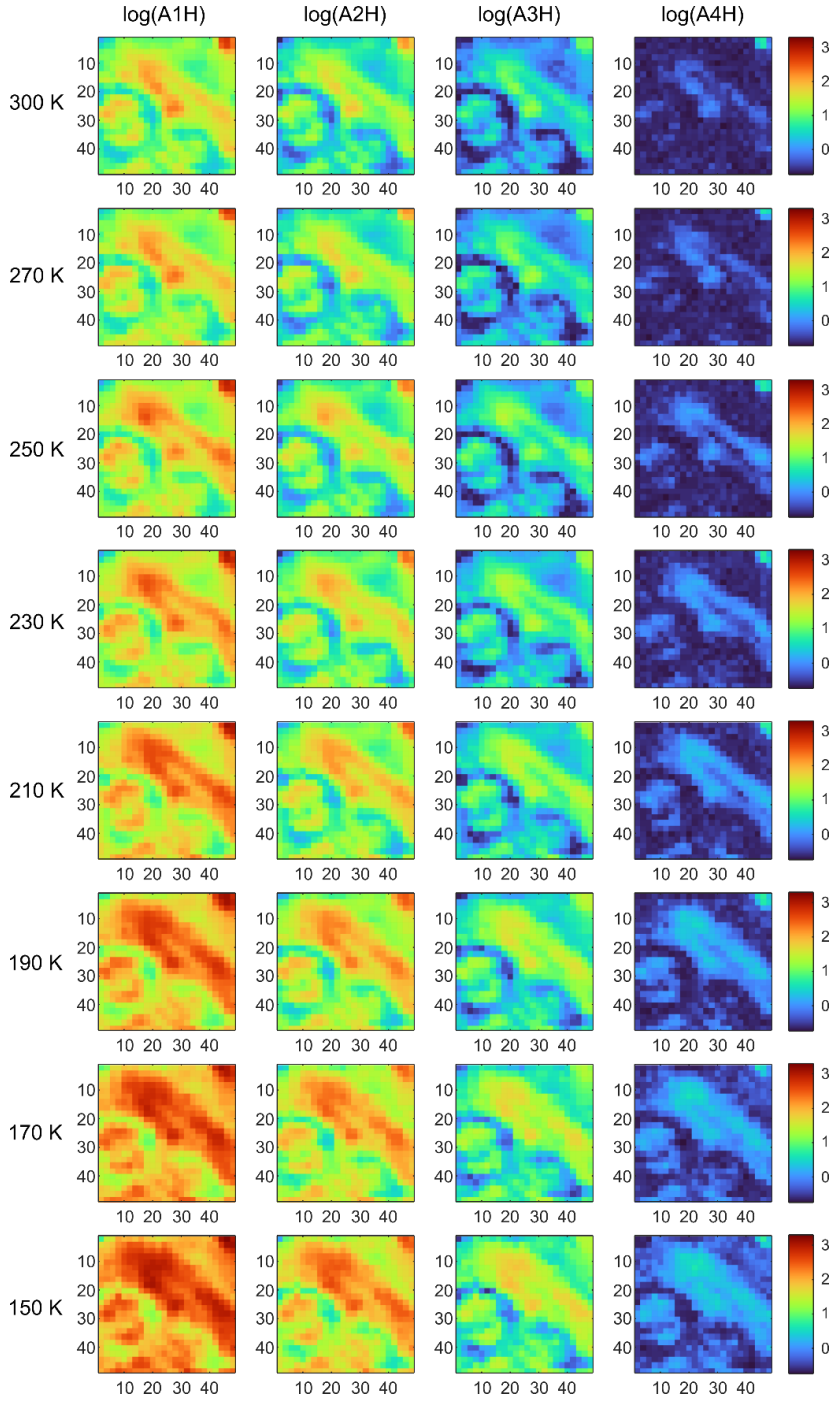
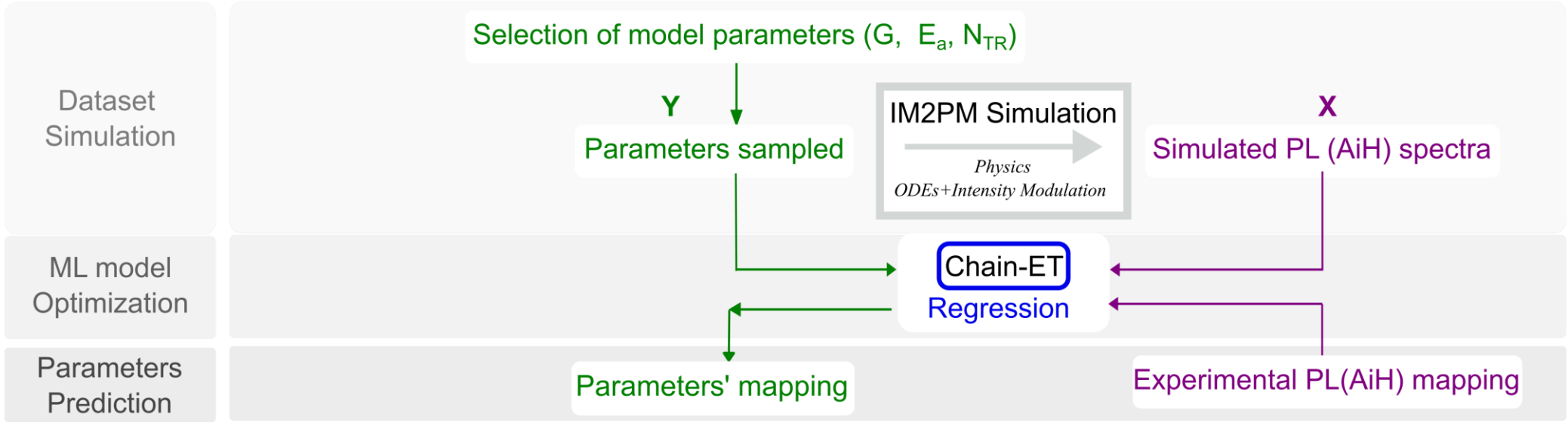


576 pixels × 32 (4 harmonics × 8 T)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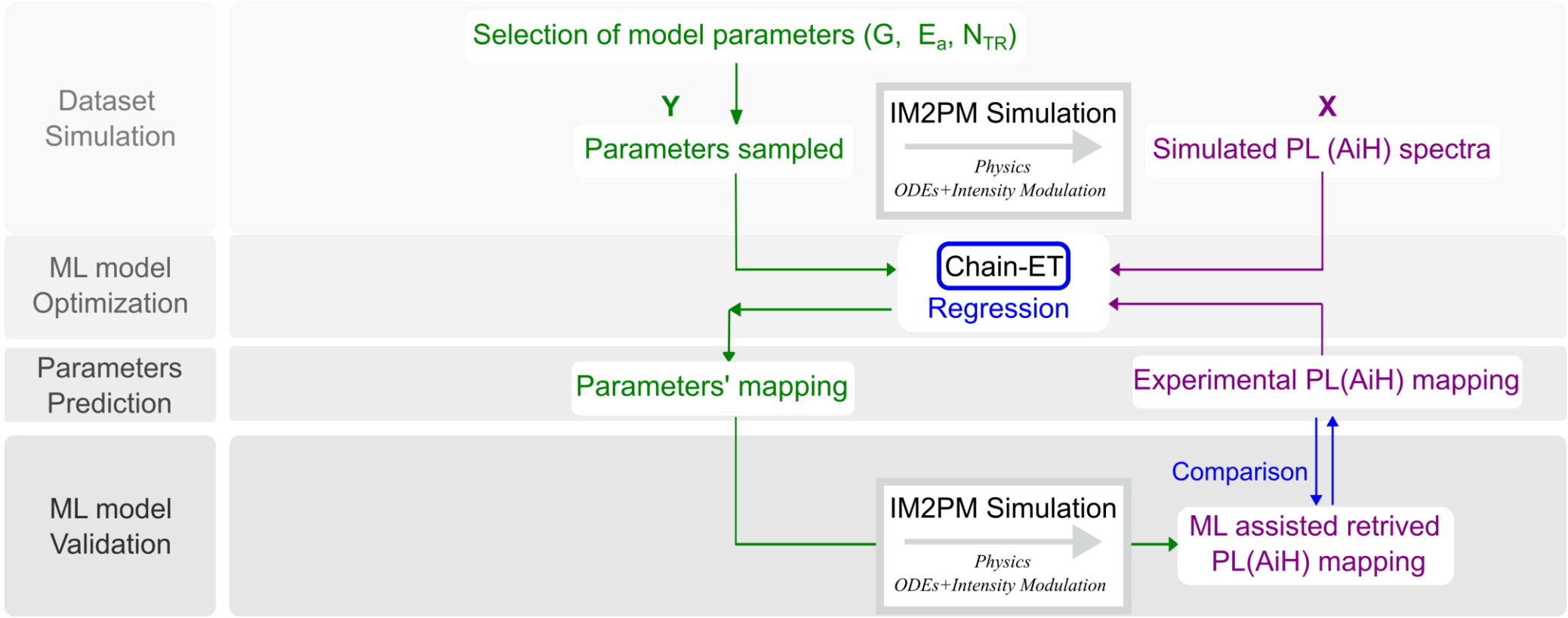
Experimental PL(AiH) mapping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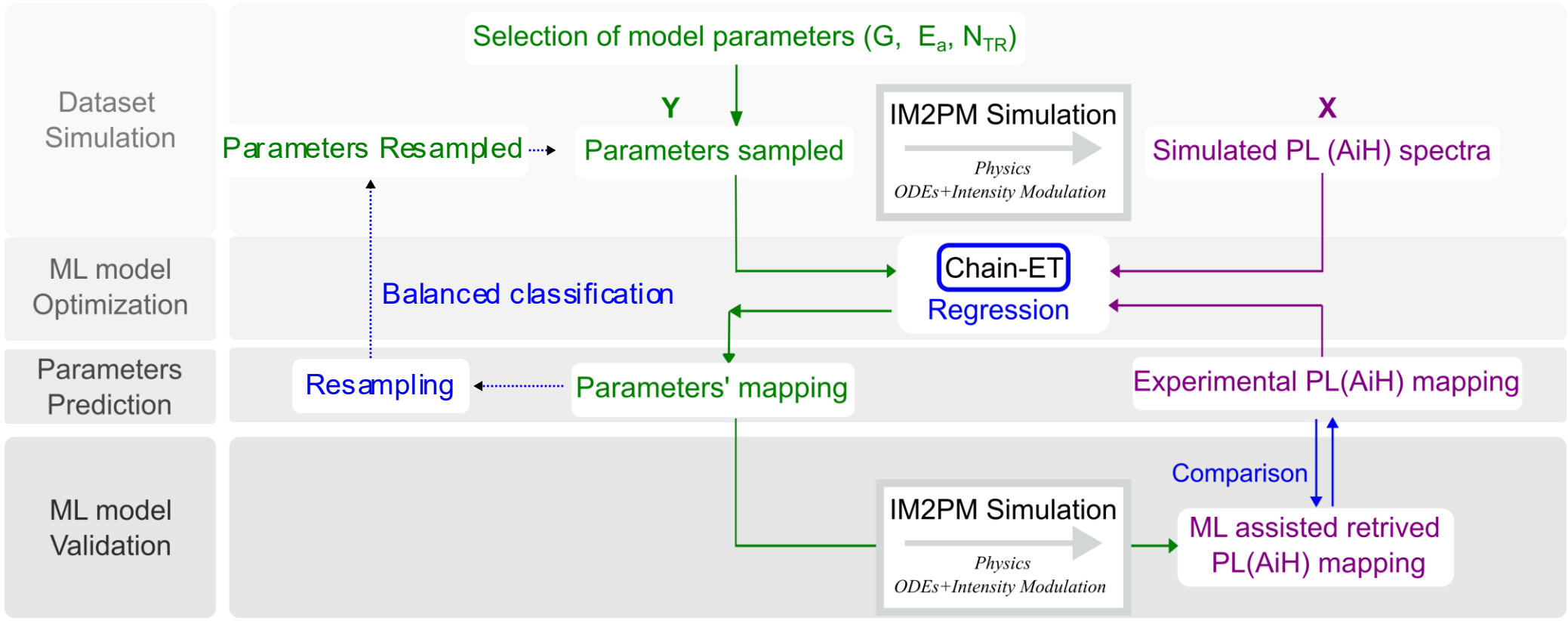
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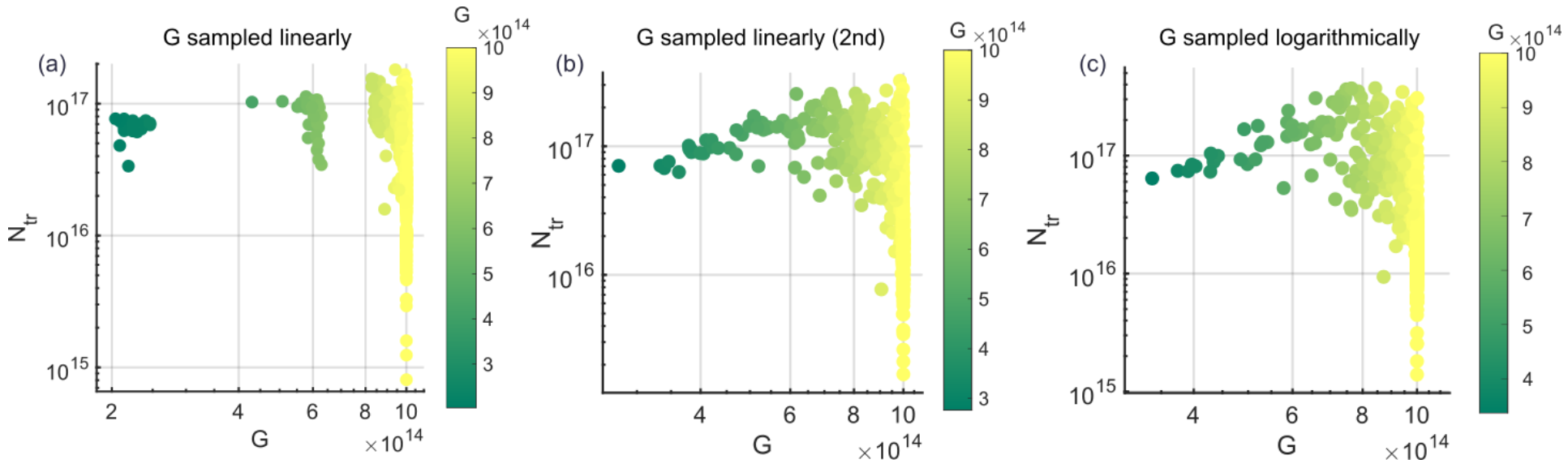
Dataset Simulation

ML model Optimization

Parameters Prediction

ML model Validation



Chain-ET		G linear	G linear 2nd value	G log
R2 coefficient	G	0.99	0.99	0.99
	NTR	0.97	0.97	0.96
	Ea	0.87	0.84	0.82
PCC coefficient Compare with Experimental data	Mean_8T	0.928	0.939	0.950
	RT	0.967	0.960	0.974

Dataset Simulation

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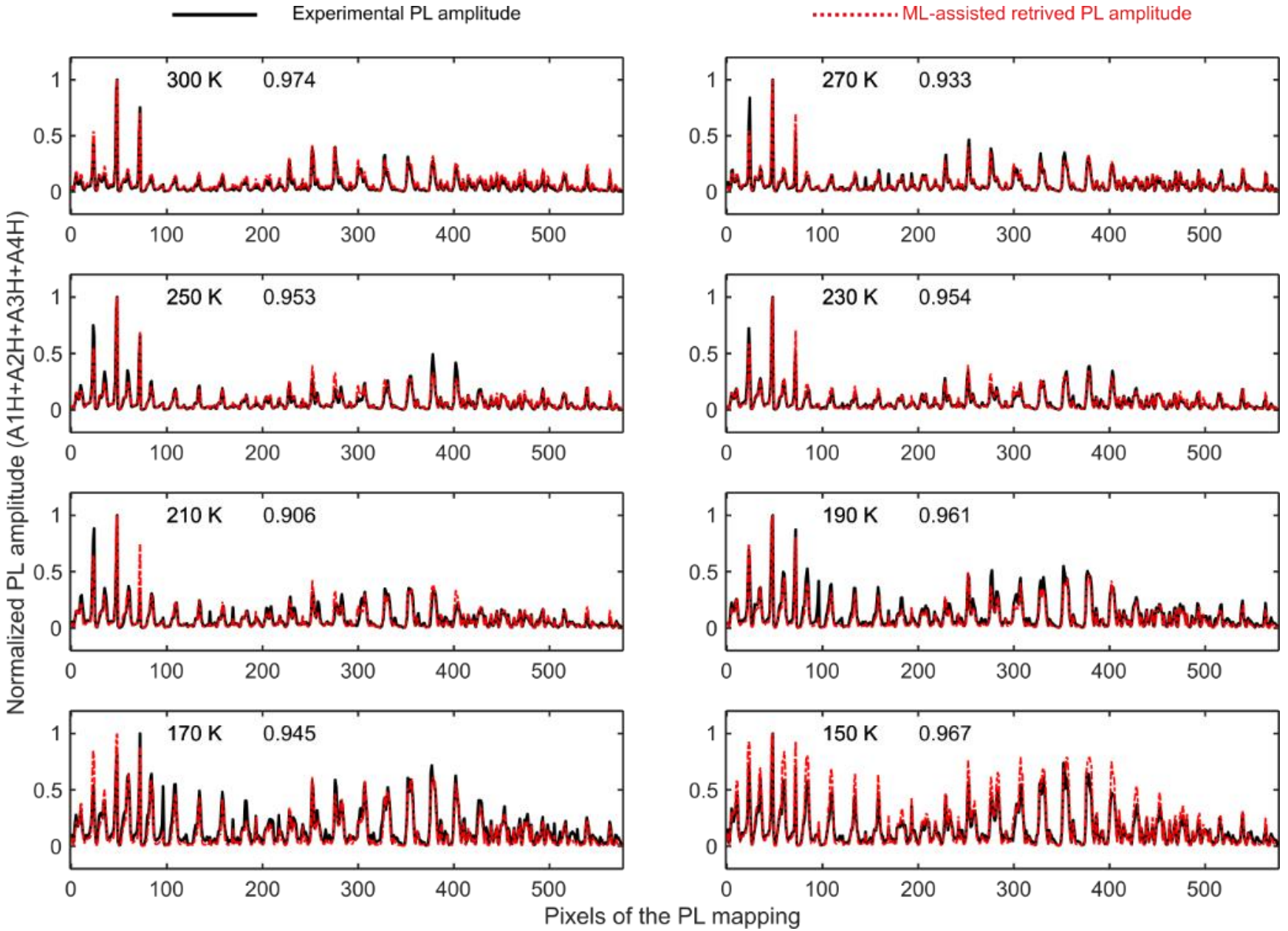
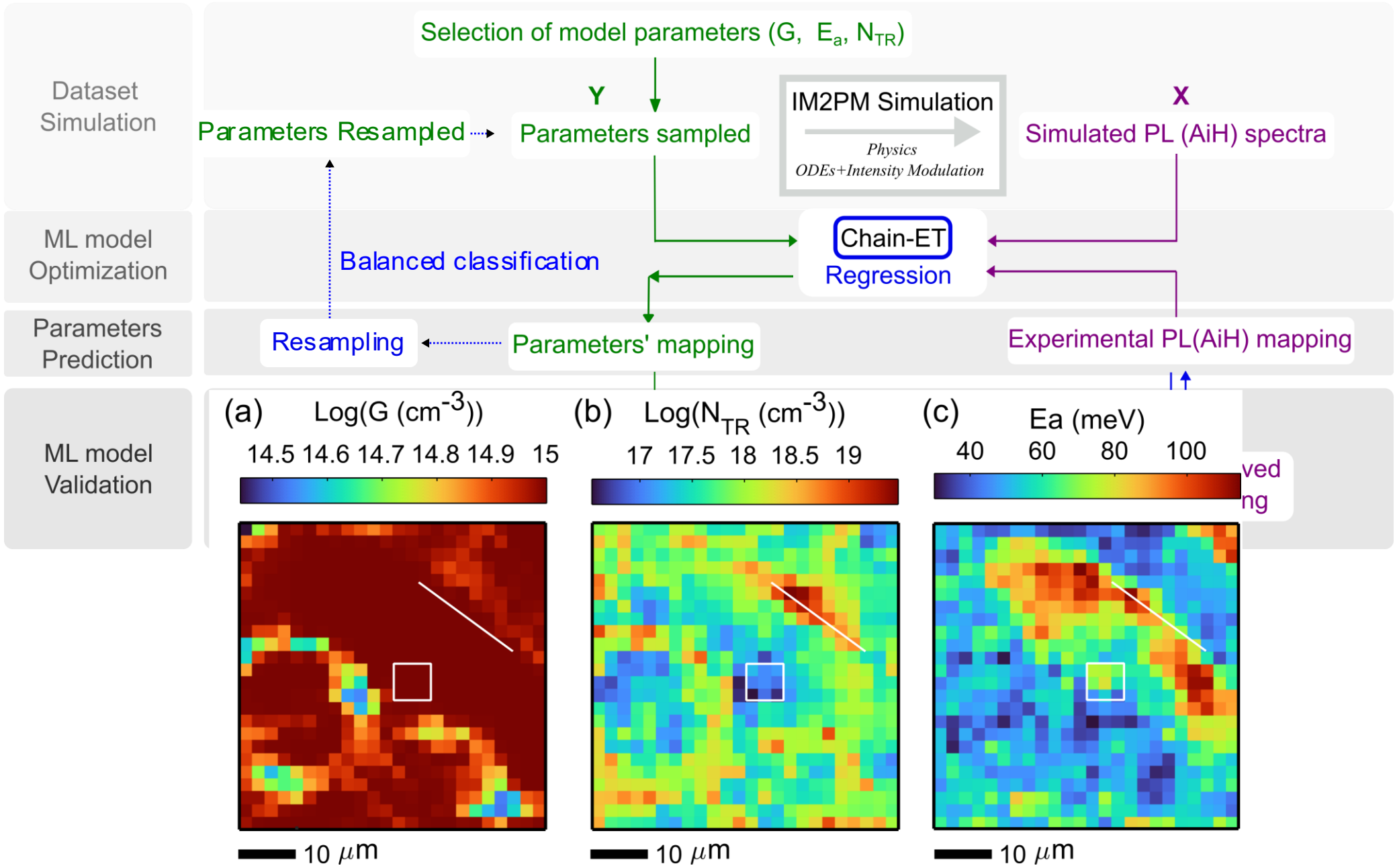
ML model Validation

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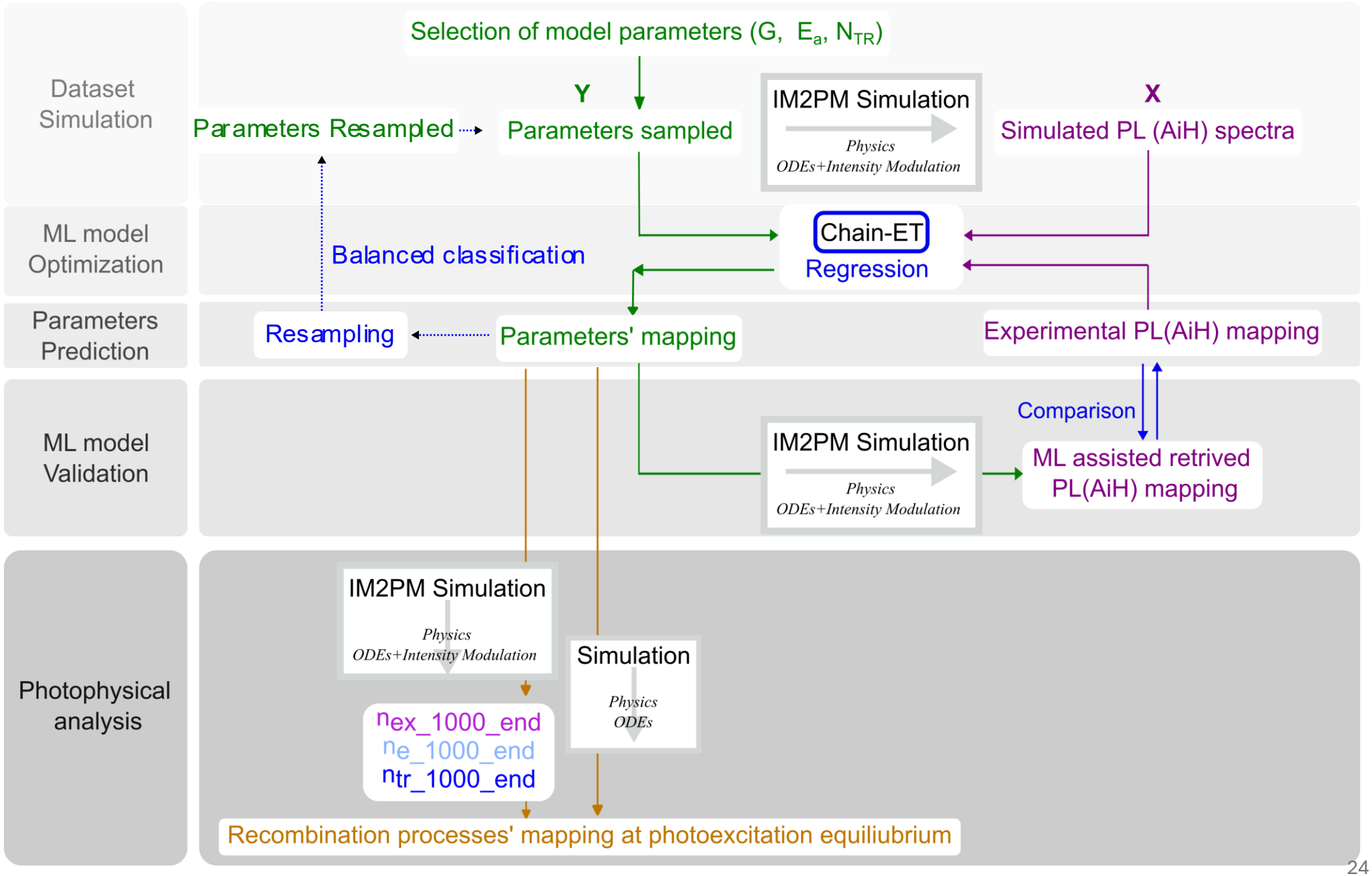
(a) Log(G (cm^{-3}))
14.5 14.6 14.7 14.8 14.9 15
(b) Log(N_{TR} (cm^{-3}))
17 17.5 18 18.5 19
(c) Ea (meV)
40 60 80 100
ved
ng
10 μm
10 μm
10 μm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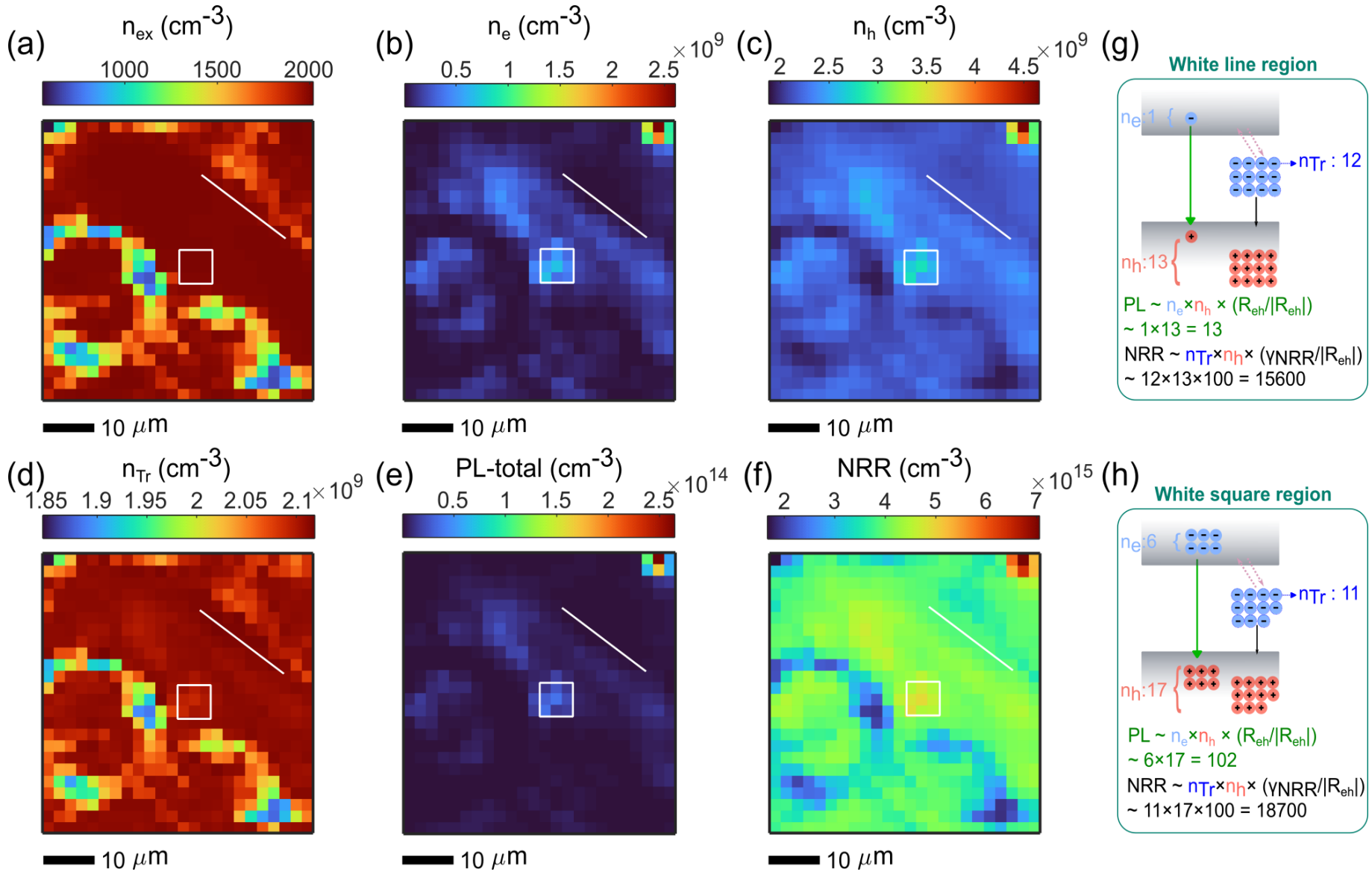
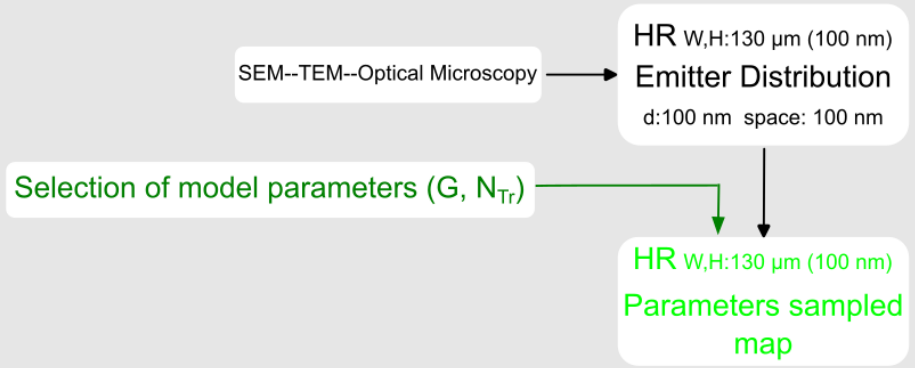
ODEs
$n_{ex\_1000\_end}$
$n_{e\_1000\_end}$
$n_{tr\_1000\_end}$
Recombination processes' mapping at photoexcitation equiliubrium


(a)
n_{ex} (cm^{-3})
1000 1500 2000
10 μm
(b)
n_e (cm^{-3})
0.5 1 1.5 2 2.5 $\times 10^9$
10 μm
(c)
n_h (cm^{-3})
2 2.5 3 3.5 4 4.5 $\times 10^9$
10 μm
(g)
White line region
n_e:1
n_{Tr} : 12
n_h:13
PL ~ $n_e \times n_h \times (R_{eh}/|R_{eh}|)$
~ 1×13 = 13
NRR ~ $n_{Tr} \times n_h \times (\gamma_{NRR}/|R_{eh}|)$
~ 12×13×100 = 15600
(d)
n_{Tr} (cm^{-3})
1.85 1.9 1.95 2 2.05 2.1 $\times 10^9$
10 μm
(e)
PL-total (cm^{-3})
0.5 1 1.5 2 2.5 $\times 10^{14}$
10 μm
(f)
NRR (cm^{-3})
2 3 4 5 6 7 $\times 10^{15}$
10 μm
(h)
White square region
n_e:6
n_{Tr} : 11
n_h:17
PL ~ $n_e \times n_h \times (R_{eh}/|R_{eh}|)$
~ 6×17 = 102
NRR ~ $n_{Tr} \times n_h \times (\gamma_{NRR}/|R_{eh}|)$
~ 11×17×100 = 18700

On going project:

$FAPbBr_3$ nanocrystals

Parameters map
High resolution



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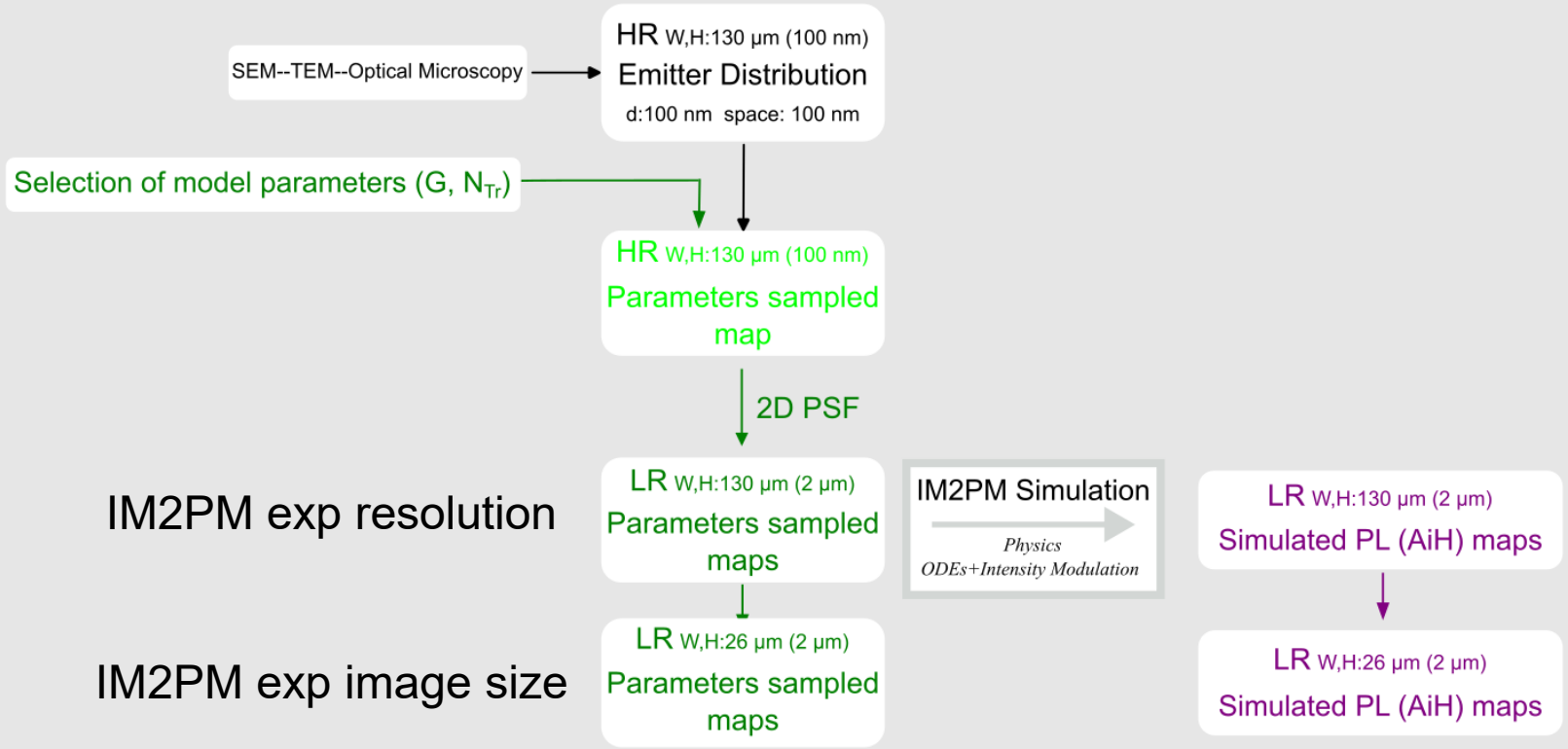
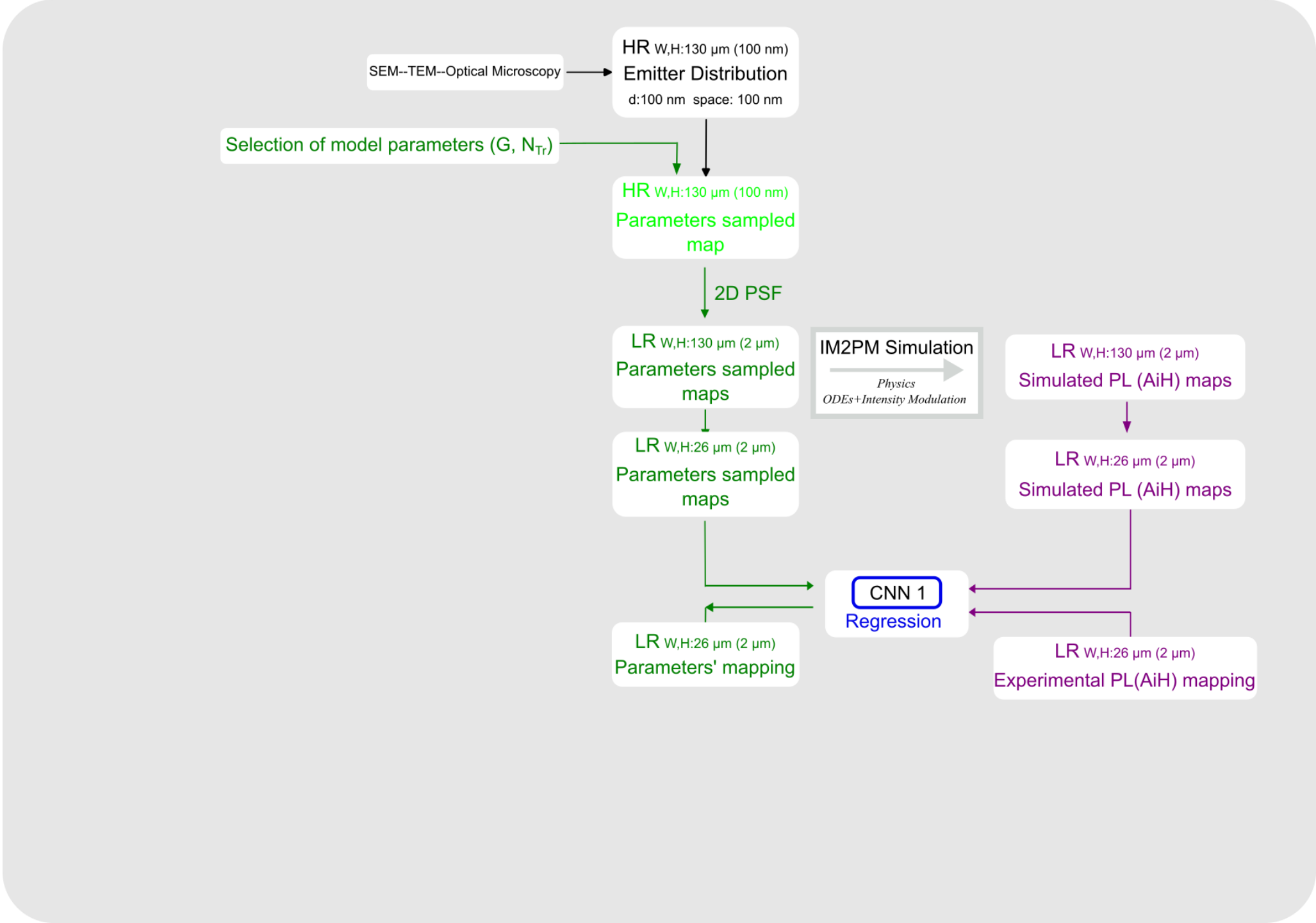
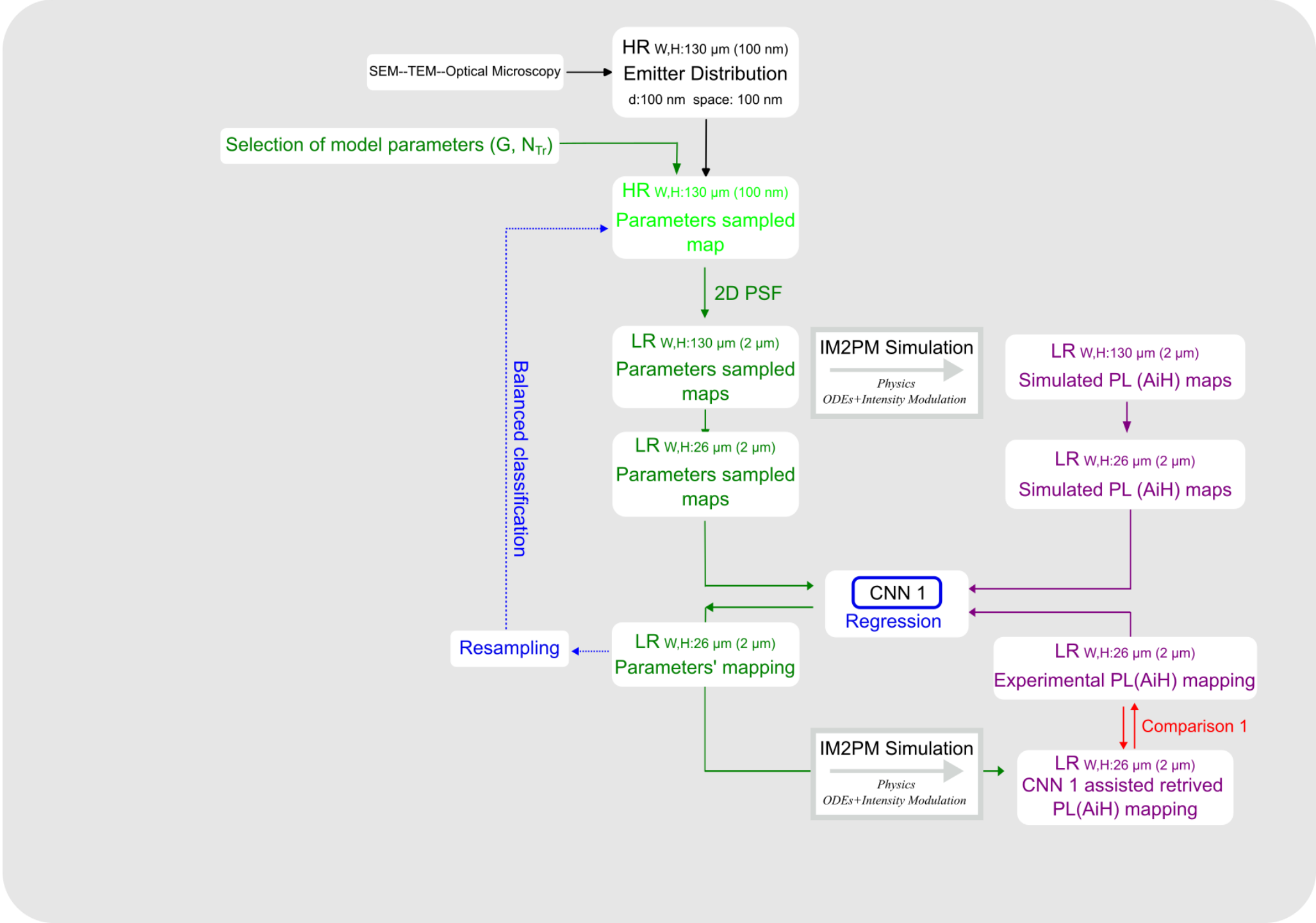
Dataset simulation



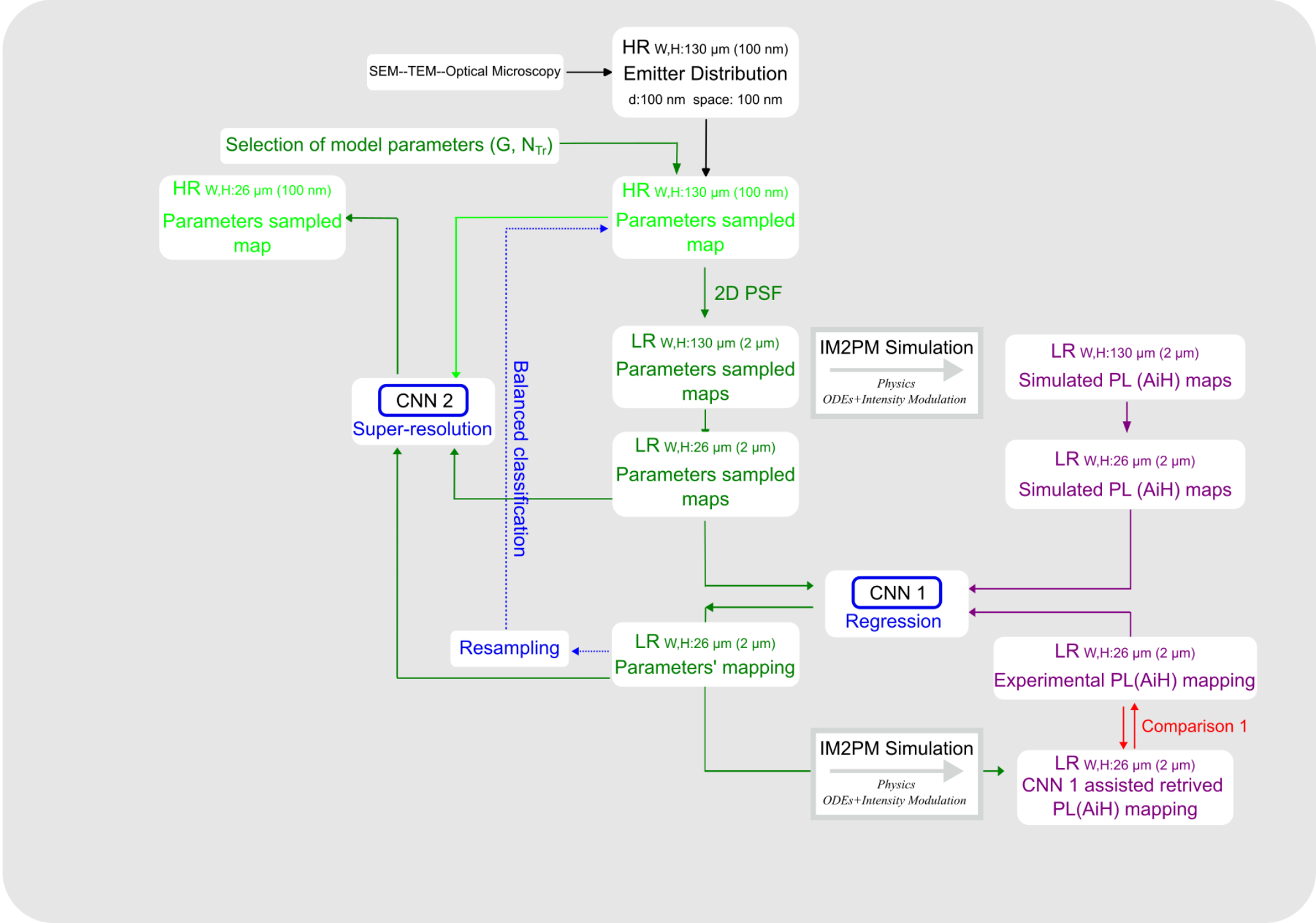
CNN1
Model optimization
Model prediction



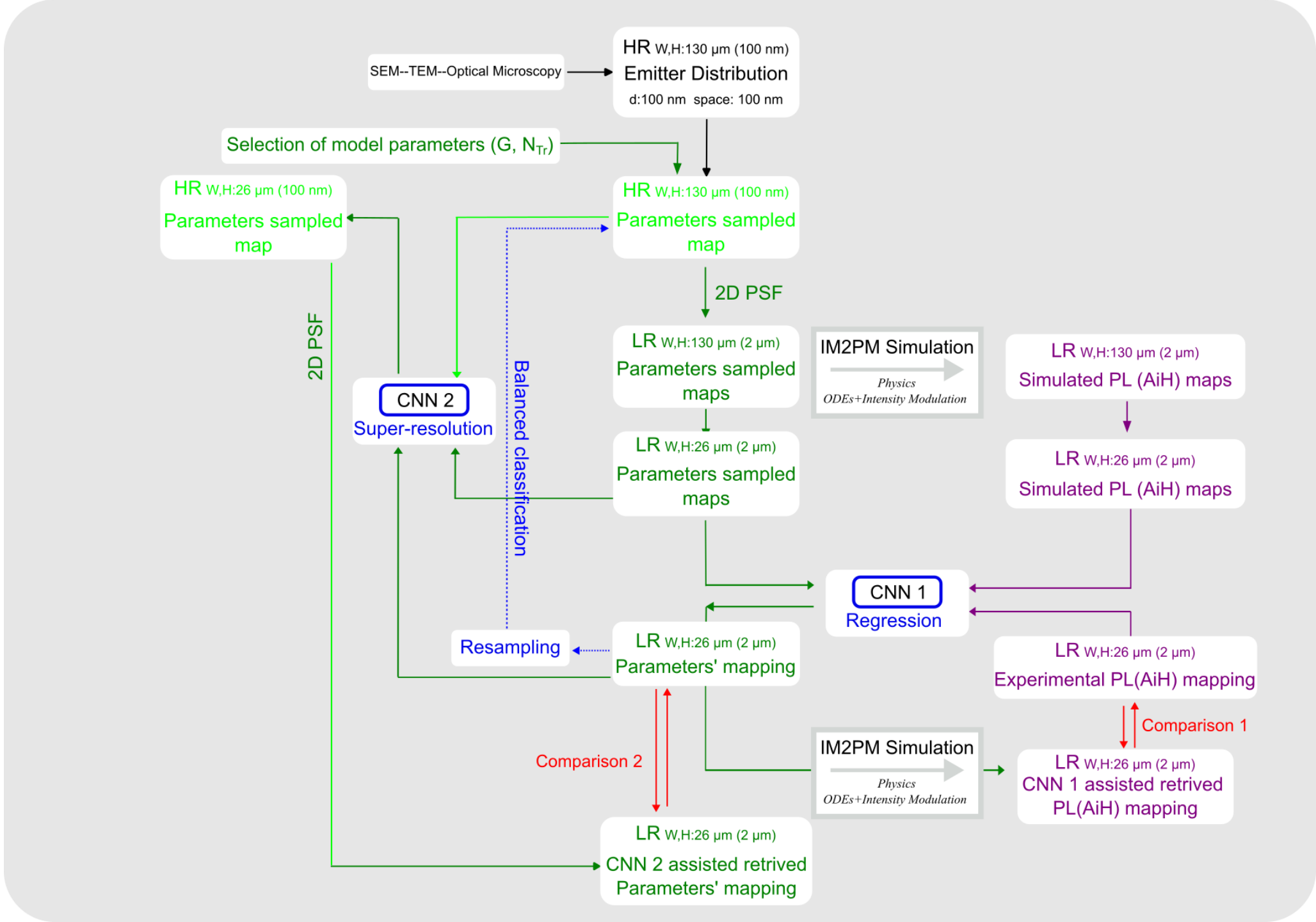
CNN1
Model validation



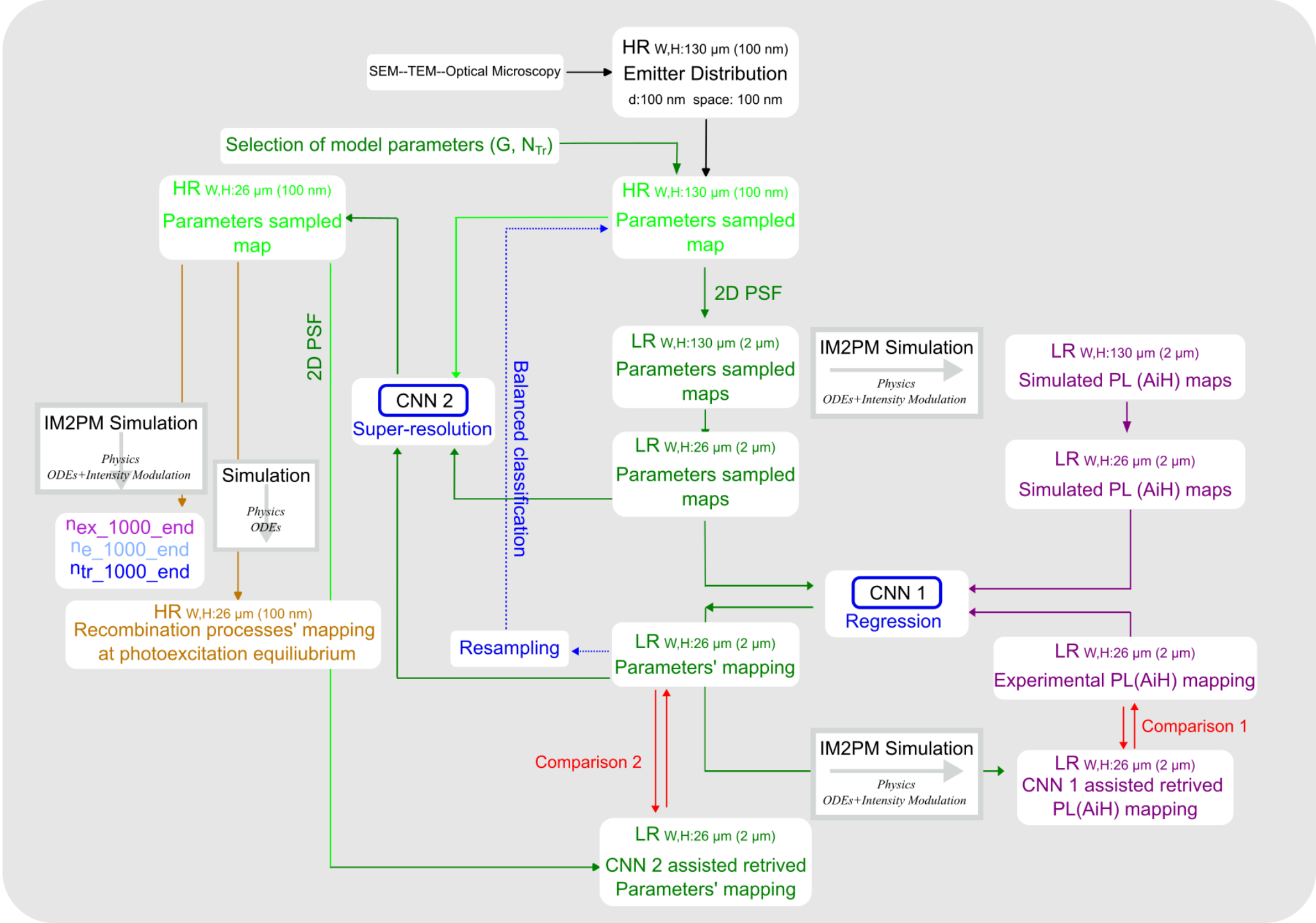
CNN2
Model optimization
Model prediction



CNN2
Model validation



High resolution
Photophysics analysis



Conclusion

- ML Regression Model for High Resolution Functional Imaging Reveals Trap States and Charge Carrier Recombination Pathways in Perovskite

- (Ongoing) Deep Learning for Super High Resolution Functional Imaging

- Thanks for your attention!