# Charting Electron Energy Loss Spectra with Machine Learning

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***Symposium CT05: Artificial Intelligence and Automation for Materials Design***

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# From the Higgs boson to quantum materials

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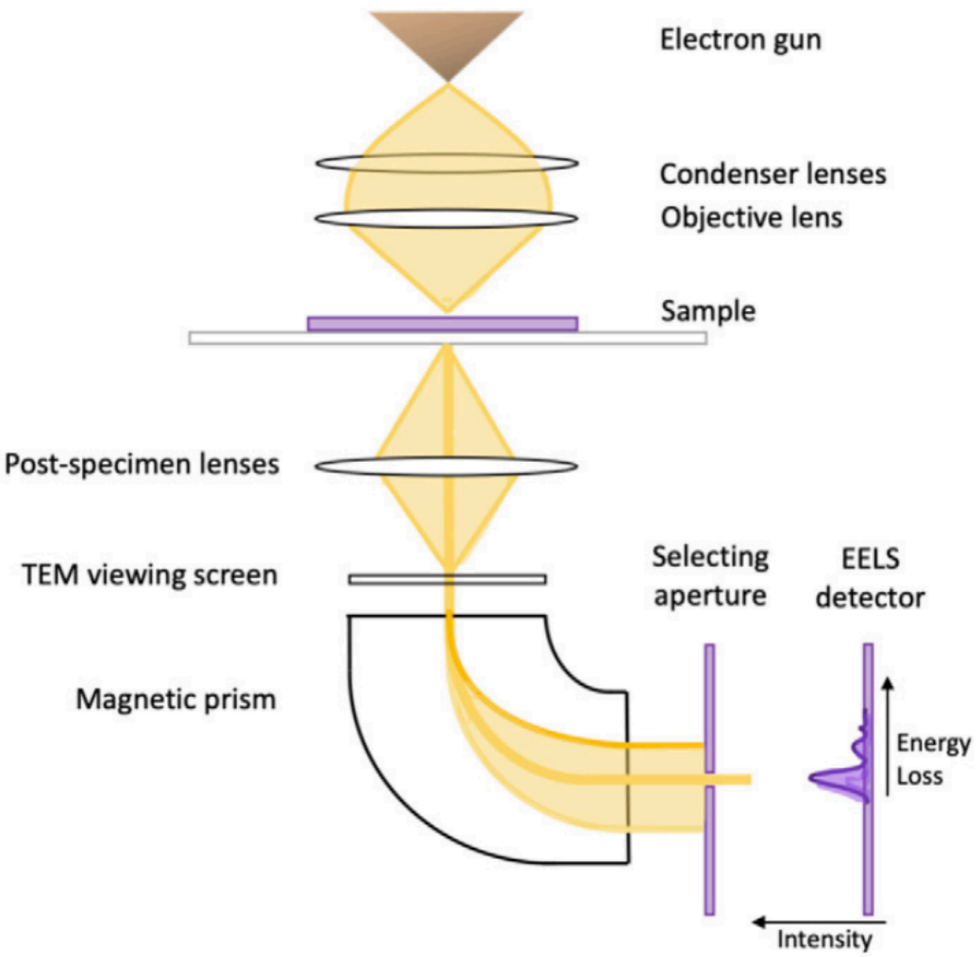
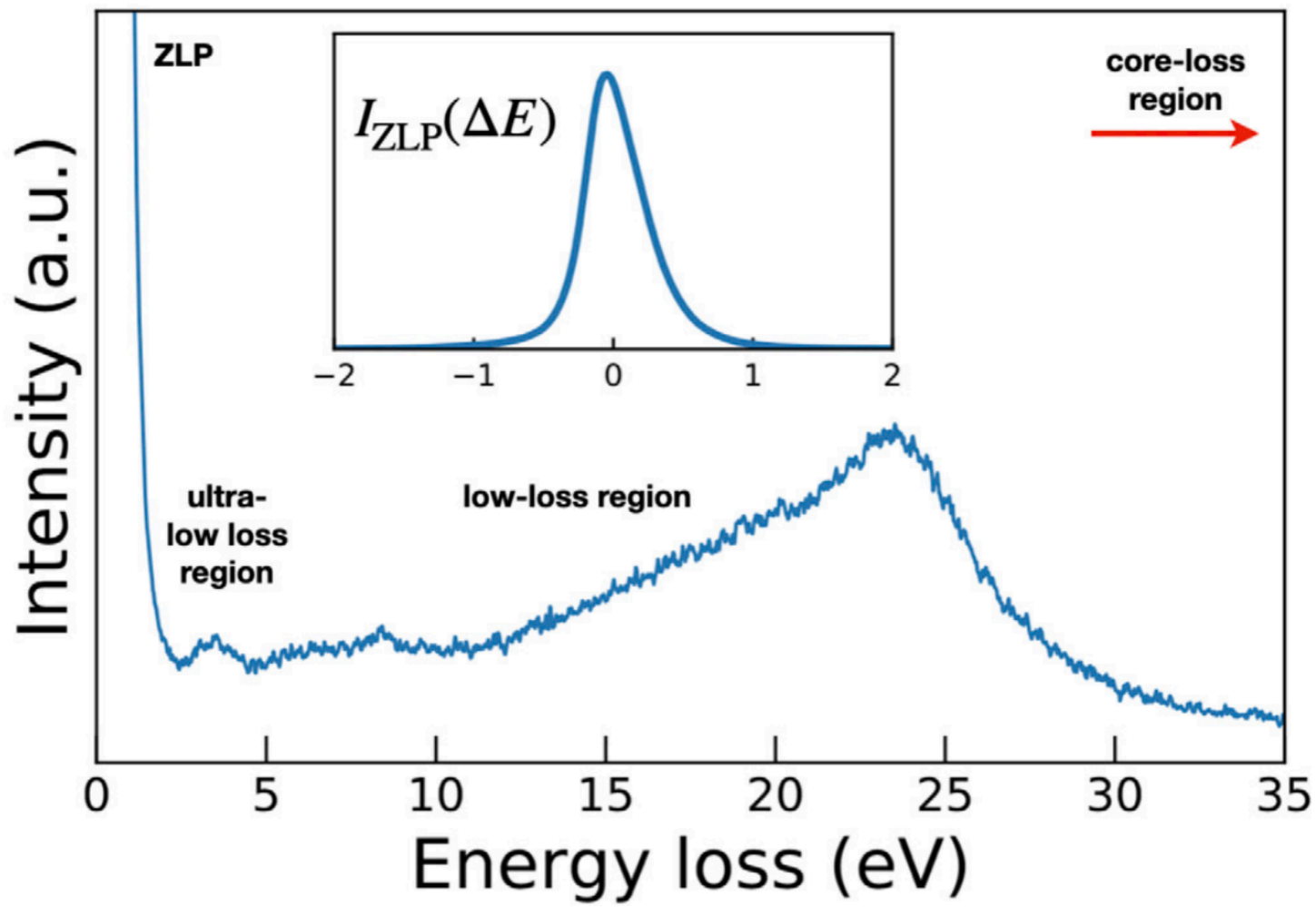
***results based on:***

- ☑ Roest, van Heijst, Maduro, Rojo, Conesa-Boj, *Ultramicroscopy* (2021)
- ☑ van Heijst, Mukai, Okunishi, Hashiguchi, Maduro, Roest, Rojo, Conesa-Boj, *Annalen der Physiek* (2021)
- ☑ Postmes, Brokkelkamp, van Heijst, ter Hoeve, Maduro, Rojo, Conesa-Boj, *in preparation*

# Electron Energy Loss Spectroscopy

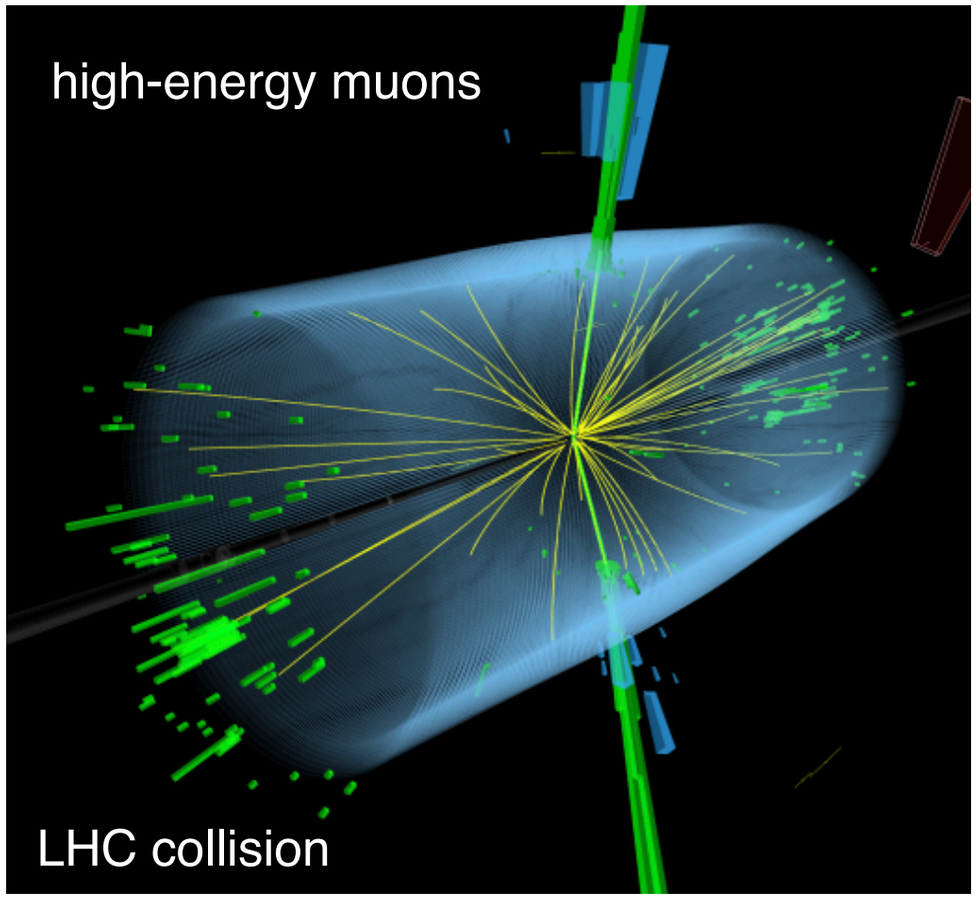
EELS: monitor **energy losses** suffered by the electrons from a Transmission Electron Microscope (TEM) beam upon **interaction with the sample**

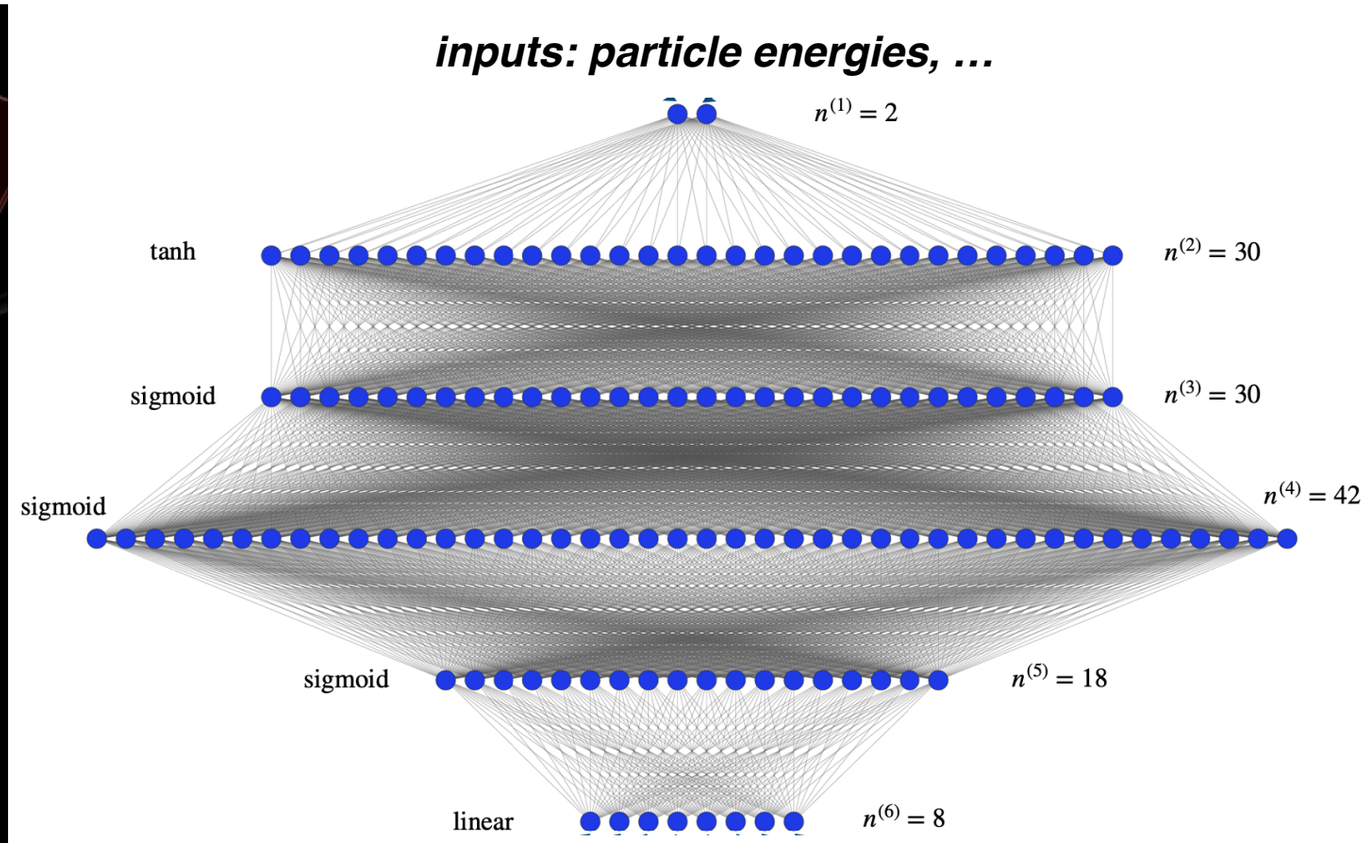


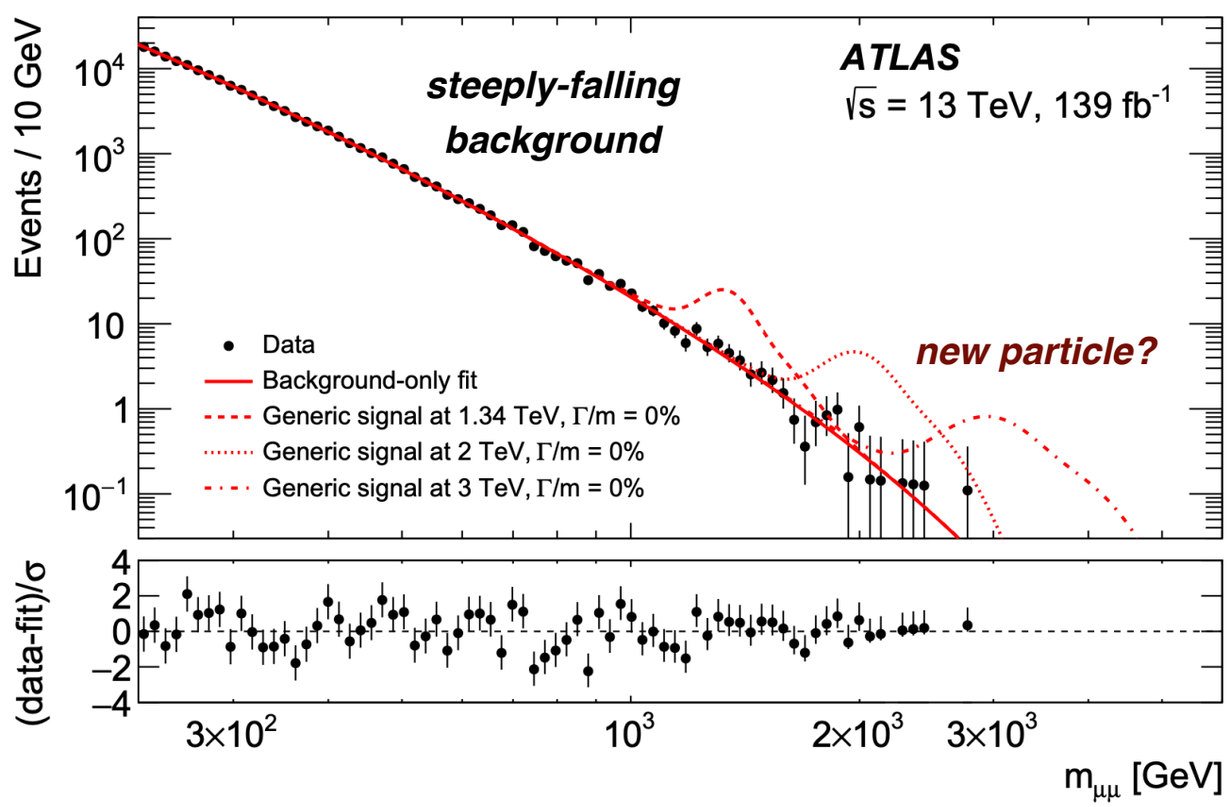




- **Challenge**: EELS measurements affected by **huge background** (zero-loss peak) at low-energy losses from elastic scatterings: complicates interpretation of material properties!
- **Solution:** parametrise backgrounds from data using **Deep Neural Networks** and **Monte Carlo sampling** to remove them in a model-independent manner

# ML-driven background subtraction in HEP

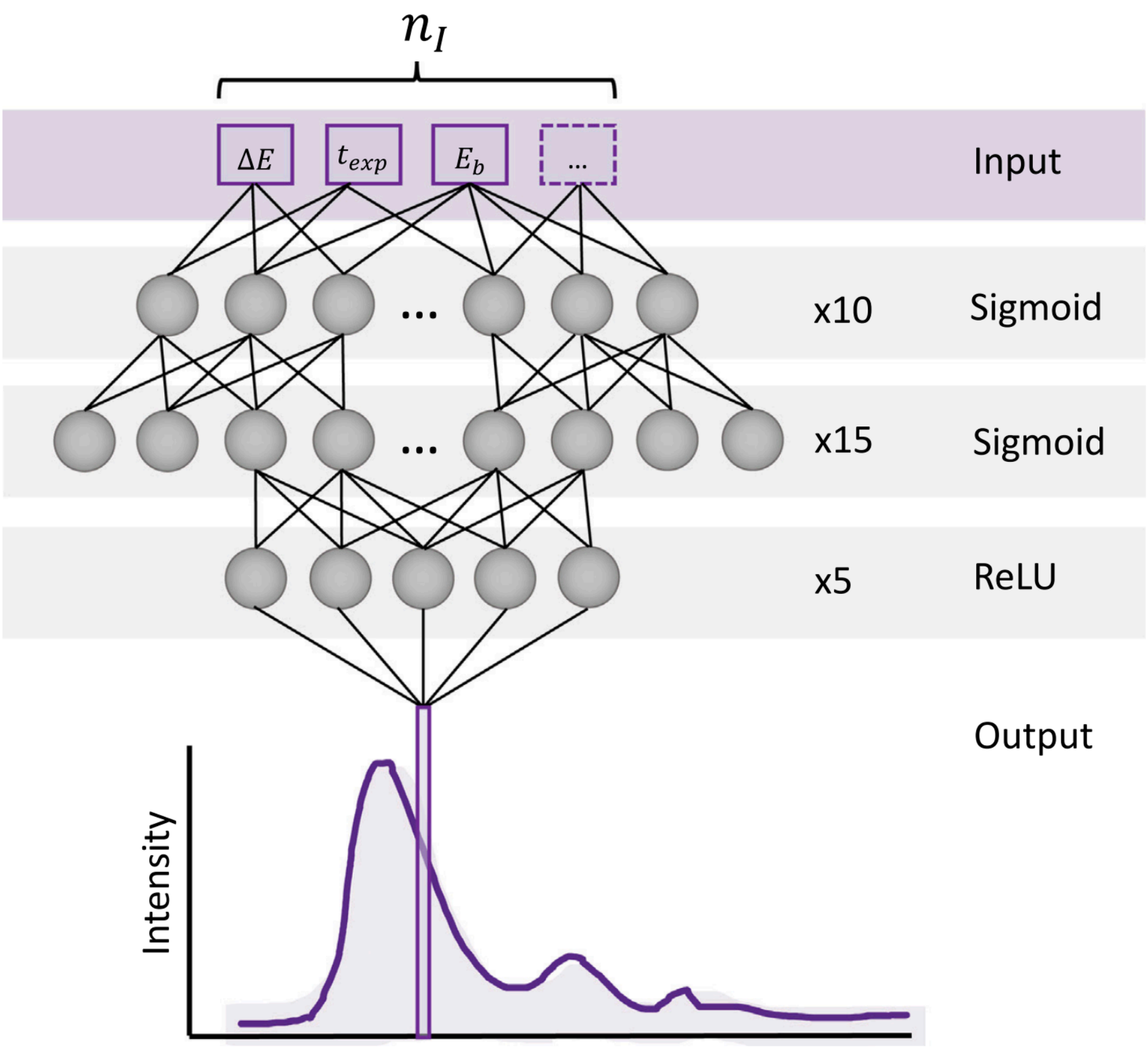



- **Learn from data** underlying physical laws and parametrise them with neural nets
- Estimate **model uncertainties** from Monte Carlo replica method: train a large number of models on *fake replicas* of actual data
- Reliable **extrapolation** to different datasets and ranges of the input variables

# A ML model for EELS backgrounds




***energy loss, beam energy, exposure time, …***

***hidden layers***

***intensity of the Zero-Loss Peak background***

**NN training performed with TensorFlow/PyTorch**

# The Monte Carlo replica method

- Generate Monte Carlo **replicas of the original data points** with multi-Gaussian distribution with central values and covariance matrices taken from the input measurements

$$I_{\mathrm{ZLP},i}^{(\mathrm{art})(k)} = I_{\mathrm{ZLP},i}^{(\mathrm{exp})} + r_i^{(\mathrm{stat},k)}\sigma_i^{(\mathrm{stat})} + \sum_{j=1}^{n_{\mathrm{sys}}} r_{i,j}^{(\mathrm{sys},k)}\sigma_{i,j}^{(\mathrm{sys})}\,, \quad \forall i\,, \quad k = 1,\ldots,N_{\mathrm{rep}}\,,$$

- Train a NN model on each replica from the **minimisation of the log-likelihood**

$$E^{(k)}\left(\{\theta^{(k)}\}\right) = \frac{1}{n_{\mathrm{dat}}}\sum_{i=1}^{n_{\mathrm{dat}}}\left(\frac{I_{\mathrm{ZLP},i}^{(\mathrm{art})(k)} - I_{\mathrm{ZLP},i}^{(\mathrm{mod})}\left(\{\theta^{(k)}\}\right)}{\sigma_i^{(\mathrm{exp})}}\right)^2,$$

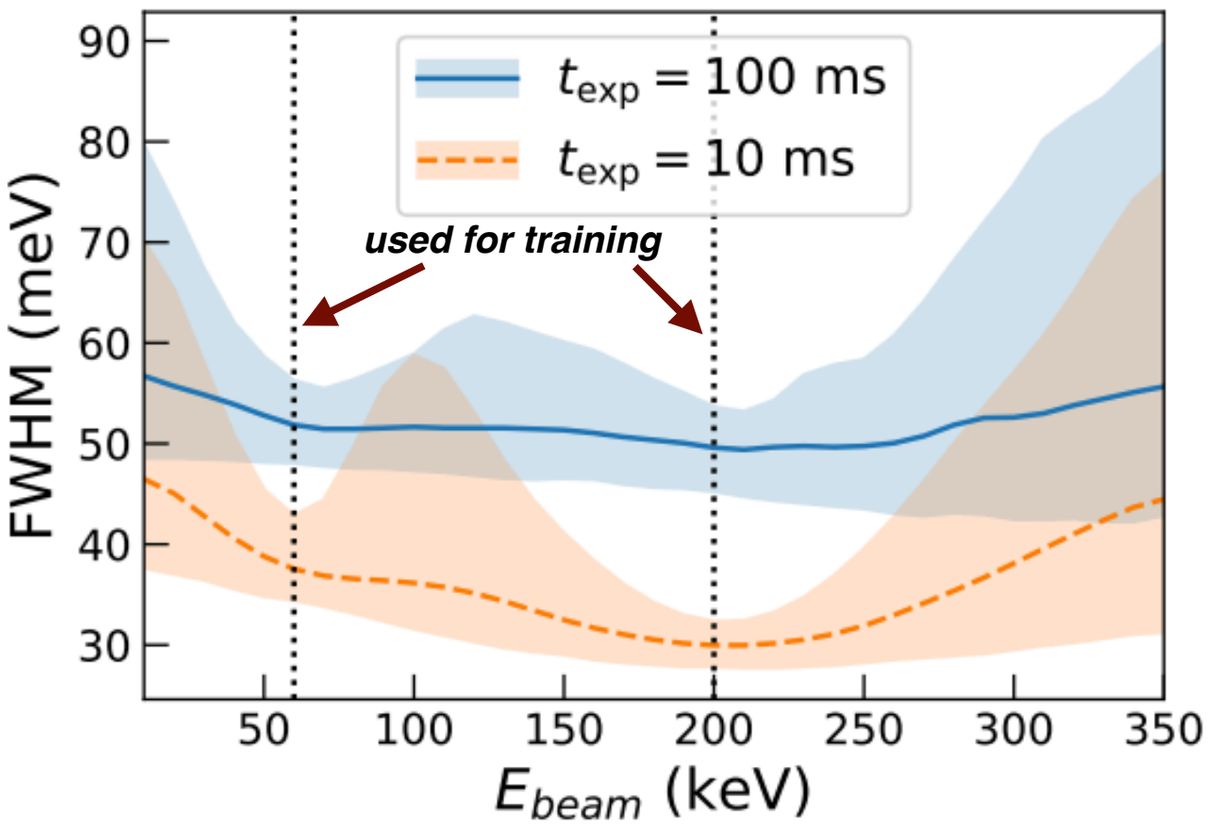
- We end up with a sampling of the **probability density in the space of NN models,** from which we can compute e.g. the variance of the predicted ZLP intensity for arbitrary inputs

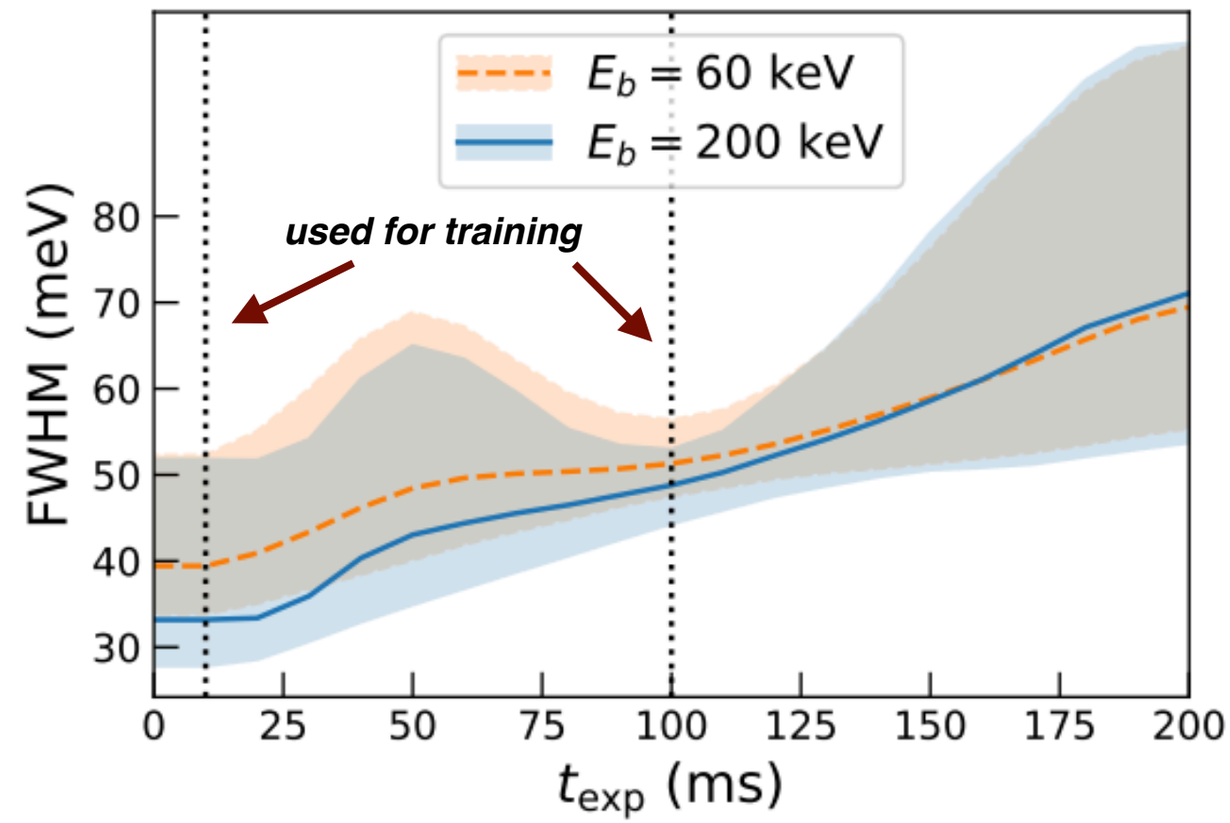
$$\sigma_{I_{\mathrm{ZLP}}}^{(\mathrm{mod})}(\{z_1\}) = \left(\frac{1}{N_{\mathrm{rep}}-1}\sum_{k=1}^{N_{\mathrm{rep}}}\left(I_{\mathrm{ZLP}}^{(\mathrm{mod})(\mathrm{k})} - \left\langle I_{\mathrm{ZLP}}^{(\mathrm{mod})}\right\rangle\right)\right)^{1/2}$$

***state of the art for error propagation in deep-learning models***

# Extrapolation to new TEM operation conditions

☑ Key property of ML: **prediction** to different ranges of input parameters



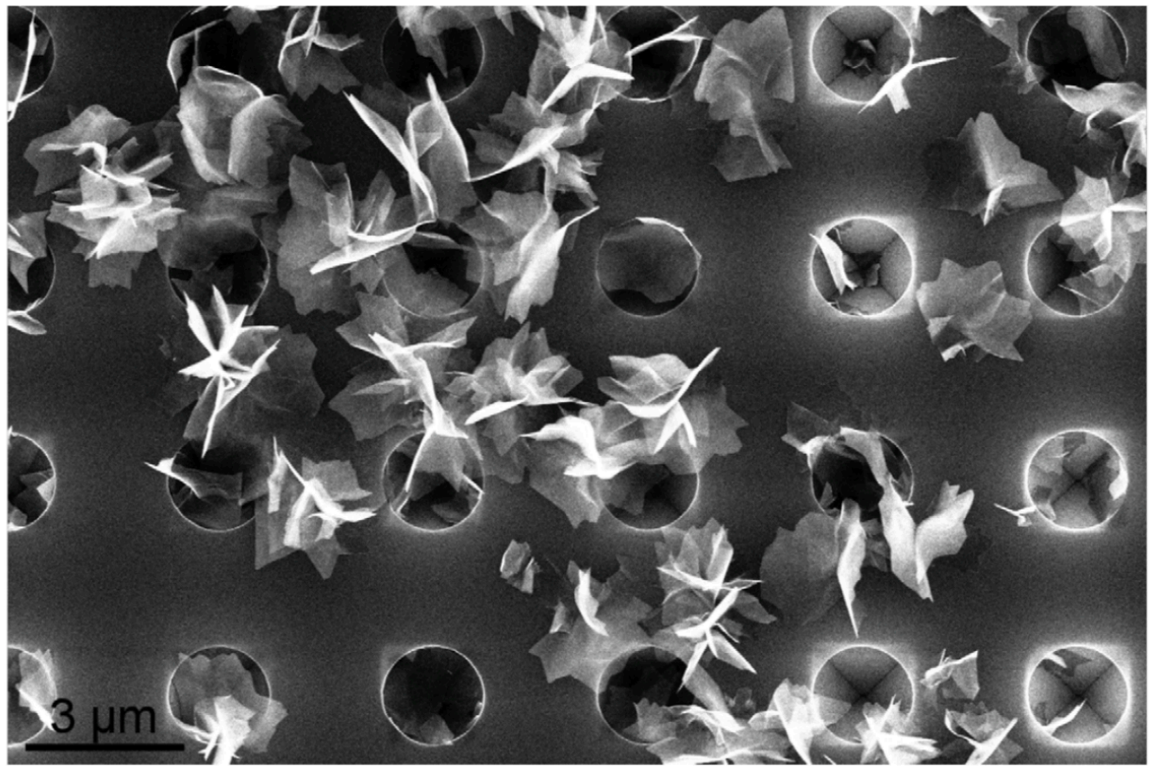



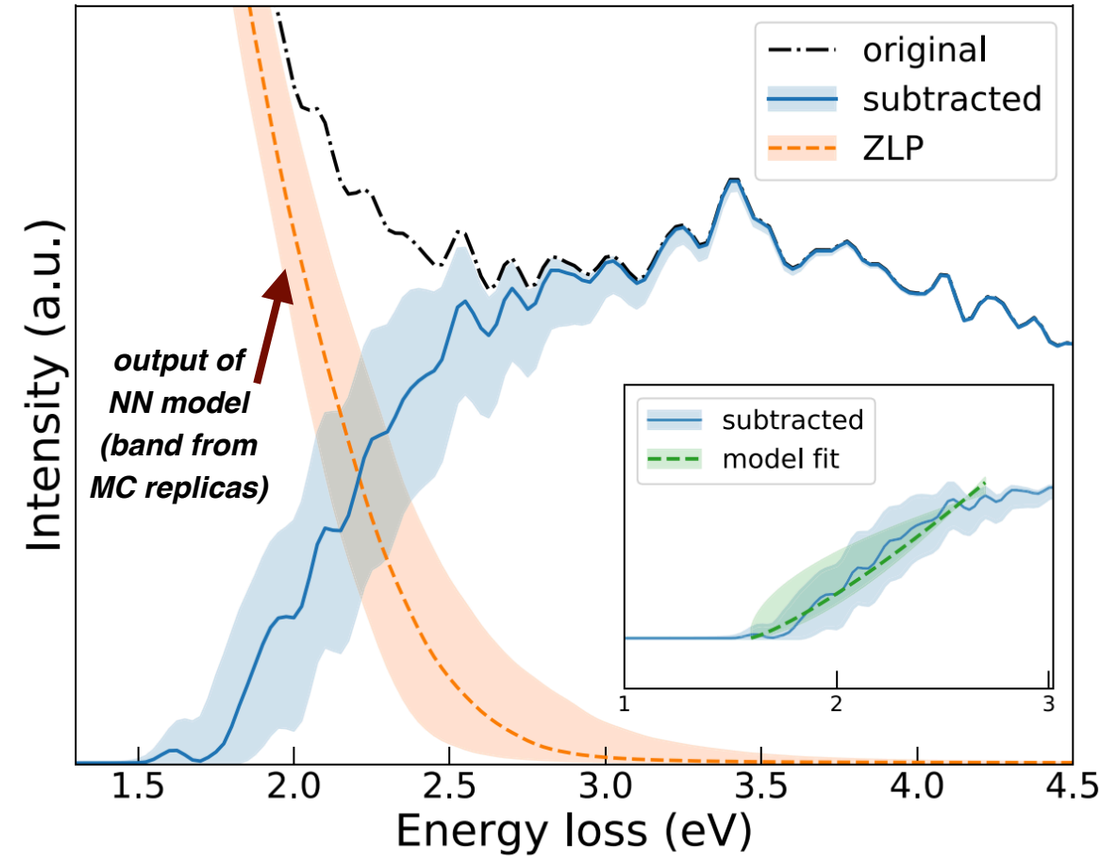

☑ Train ZLP model for specific values of TEM operation parameters, e.g. **electron beam energy** and then inter/extrapolate outside training range

☑ The model **uncertainties increase** when our prediction is not reliable and more data needed

☑ Important: **no assumptions** of functional dependence of background model with input variables

# Band gap extraction in polytypic $WS_2$



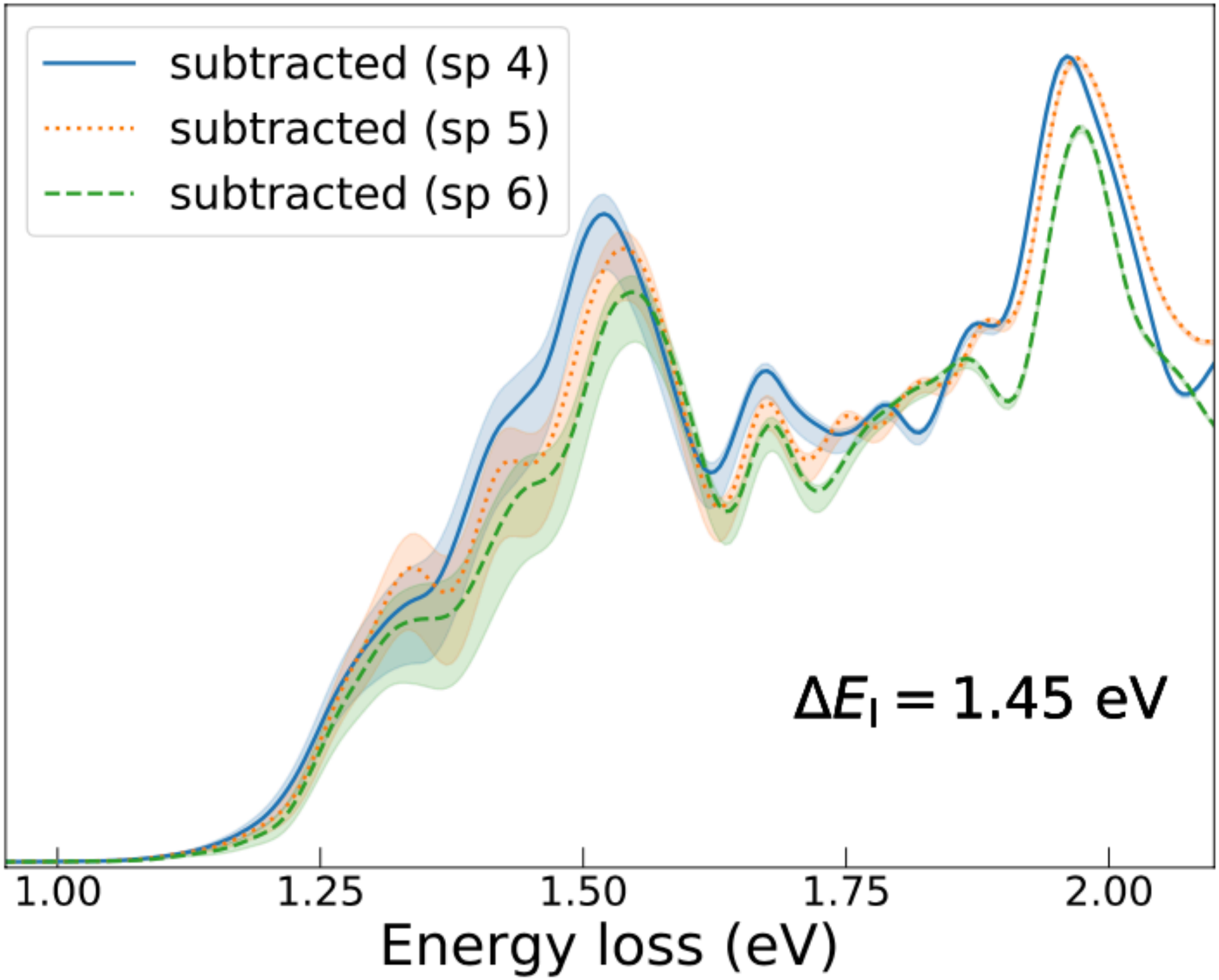




- ☑ Apply to **nanoflowers** composed by **polytypic $WS_2$** (a 2D quantum material)
- ☑ First extraction of band gap in this material from fit to subtracted EEL spectra

$$I_{\text{inel}}(\Delta E) \simeq A\,(\Delta E - E_{\text{BG}})^b \qquad E_{\text{BG}} = 1.6^{+0.3}_{-0.2}\,\text{eV}\,, \qquad b = 1.3^{+0.3}_{-0.7}\,.$$

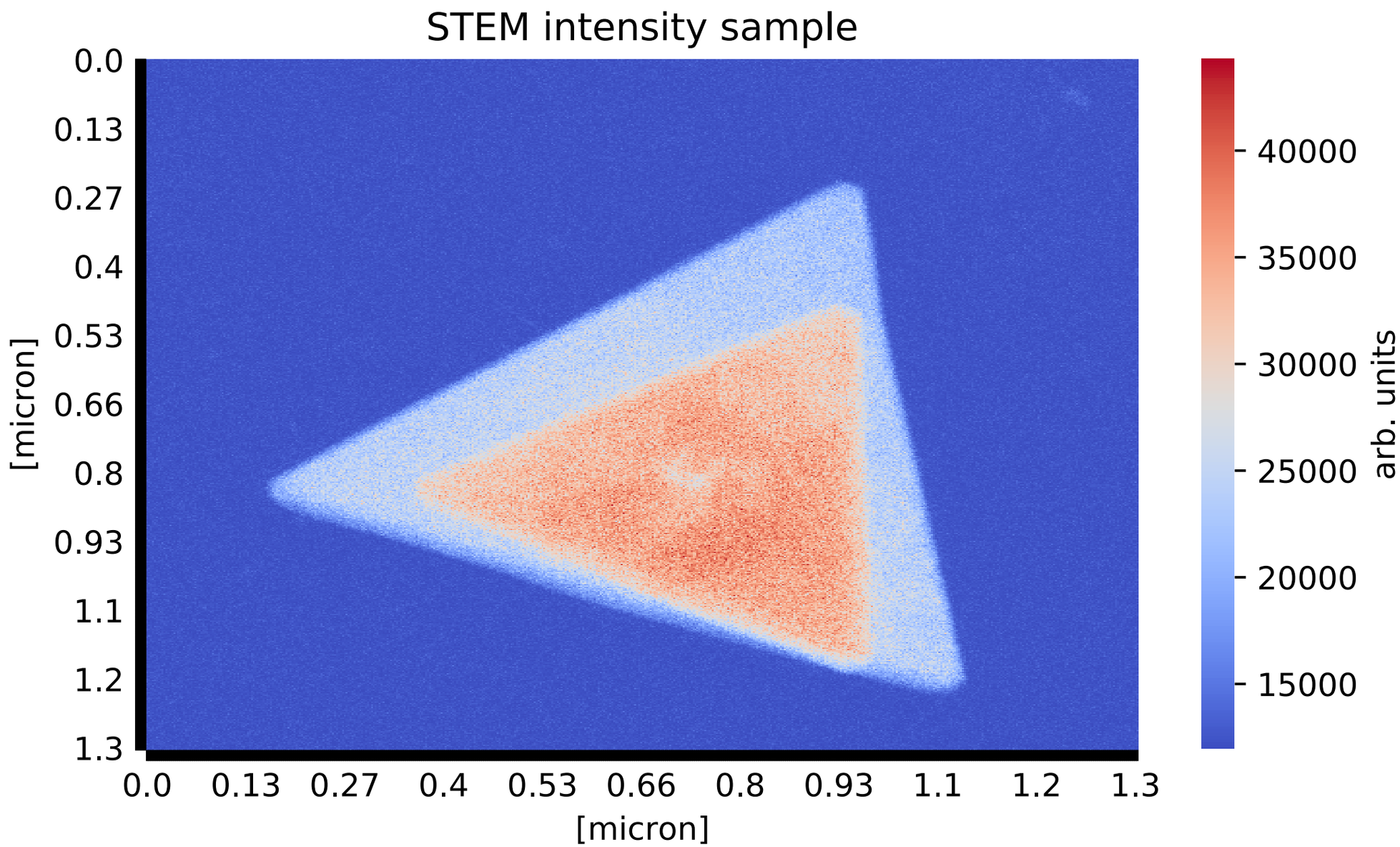
- ☑ ML-subtracted spectra make possible mapping **exciton transitions** down to 1.5 eV

# Band gap extraction in polytypic $WS_2$




☑ ML-subtracted spectra make possible mapping **exciton transitions** down to 1.5 eV

# ML analysis of spectral images




- ☑ EELS spectral image contains up to **O($10^5$) individual spectra**
- ☑ Use **unsupervised learning** (*K-means clustering*) to identify clusters of pixels with comparable sample thickness and combine them for the **(supervised) NN training**
- ☑ Simultaneous determination of physical properties across the **whole nanostructure** with their **uncertainties:** thickness, band gap, position and width of plasmonic and excitonic resonances,…

# Summary and outlook

**Machine learning algorithms** offer exciting avenues to improve and boost **data interpretation** in electron microscopy and related techniques

- ☑ The combination of **deep learning models** and **Monte Carlo replica methods** make possible assumption-free, faithful background subtraction in EELS spectra

- ☑ One can reliable predict the shape and magnitude of these backgrounds **for other operation conditions** beyond those used in the training

- ☑ The methodology can be applied to any other problem where **large (multi-dimensional) backgrounds needs to be removed** in order to access the relevant physical information

results obtained with **EELSfitter** code, publicly available in GitHub:

***https://github.com/LHCfitNikhef/EELSfitter***