A Critical Review of Neural Networks for the Use with Spectroscopic Data

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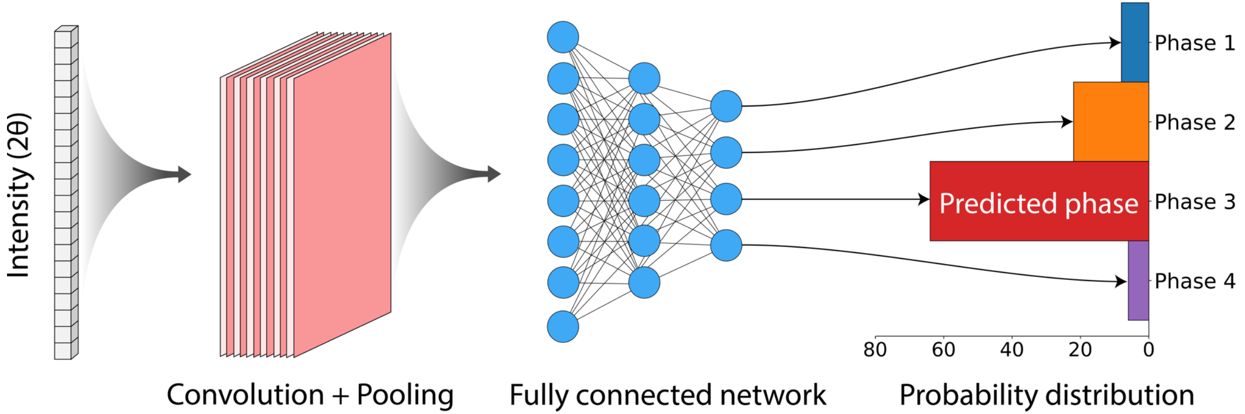
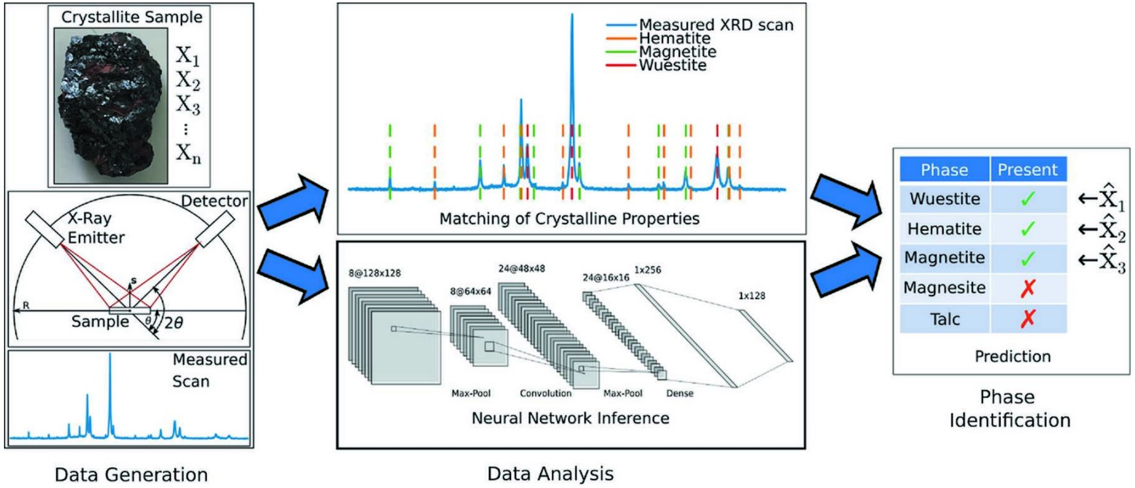


Outline

- Introduction
- Related Work
- Evaluation Dataset
- Recent Developments
- Conclusion

Introduction - Topic

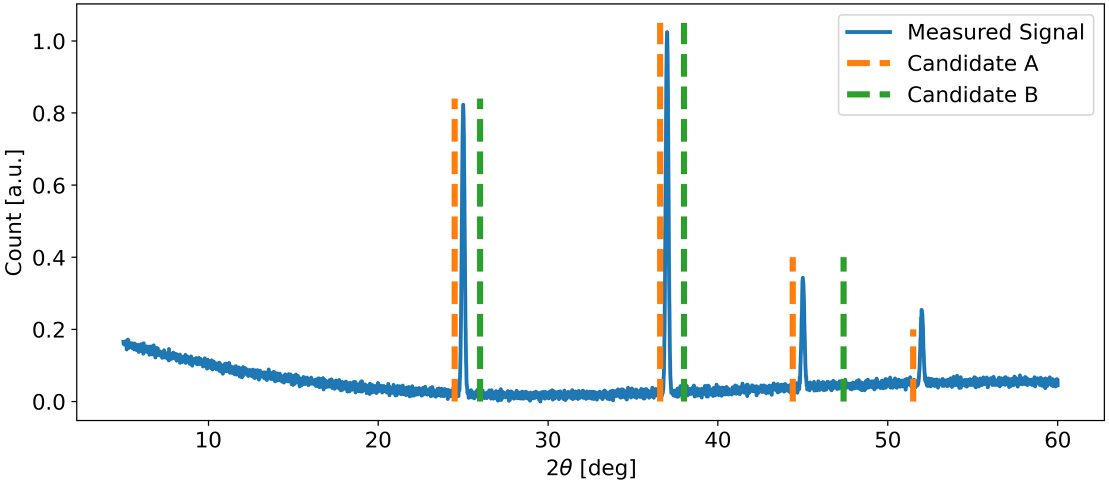
- Machine learning (ML) methods popular for spectra analysis
- Neural networks used for X-ray diffraction (XRD), Raman spectroscopy, etc.
- E.g., XRD 1D powder spectra → typical task: phase identification



From Schuetzke et al. 2021 [1]

Introduction - Challenges

- Matching measured intensities with references "pattern matching" → classification task
- Picking candidates based on peak positions and intensities
- Variation of positions, intensities, shapes, background, etc.



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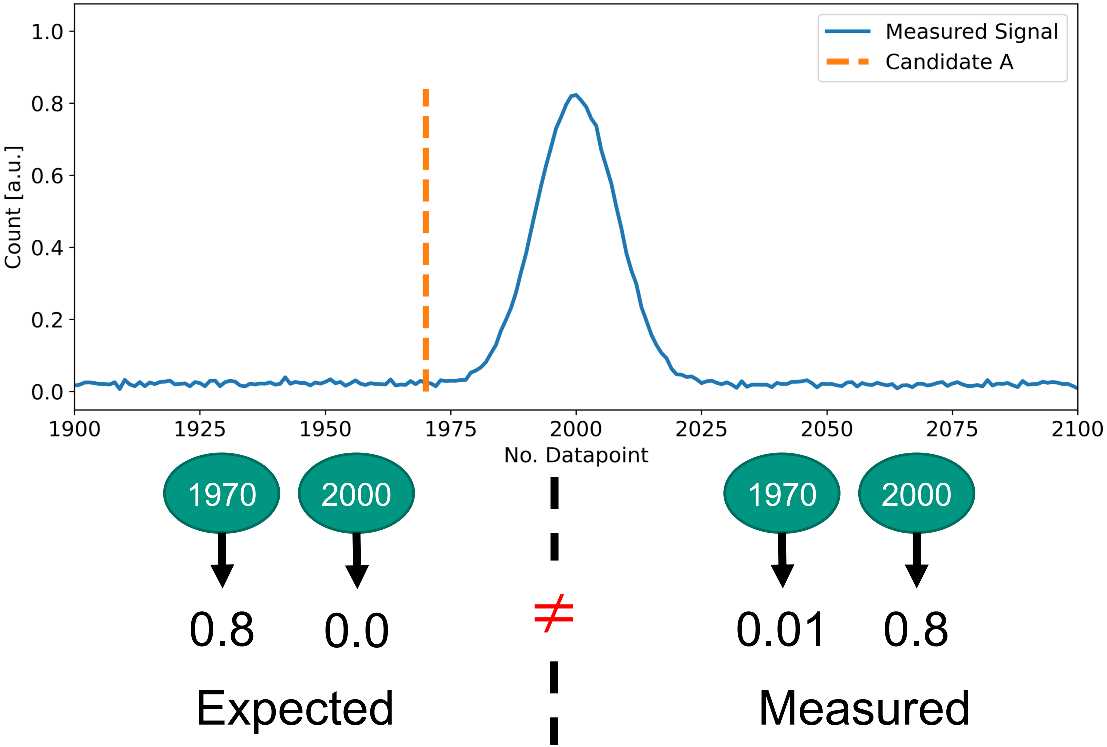
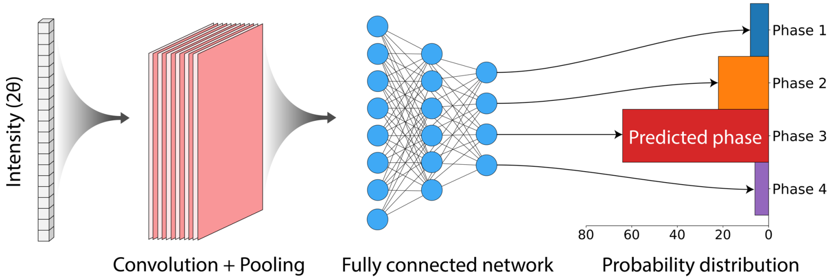
Introduction – Machine Learning Models

- Machine Learning models learn thresholds per dimension
- For spectra: each datapoint a separate dimension
- Problem with <u>shifts</u>: various dissimilarity metrics to account for position variation [2]



Related Work – Neural Networks for Spectra

- Neural Network models applied to spectroscopic data of various domains; improvement over traditional ML models
- Models mostly use Convolutional Neural Network (CNN) structure
- BUT no network achieved perfect prediction accuracy in recent benchmark study [6]

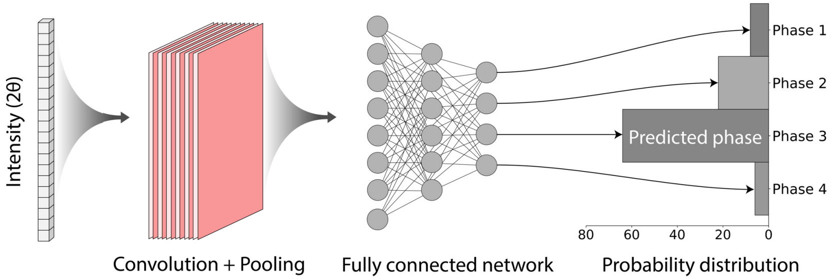


From Szymanski et al. 2021 [7]

Publication	Type	Architecture
Liu et al., 2017 [3]	Raman	3 Convolutional Layers
Cui and Fearn 2018 [4]	Near-infrared	1 Convolutional Layer
Lee et al., 2020 [5]	XRD	3 Convolutional Layers

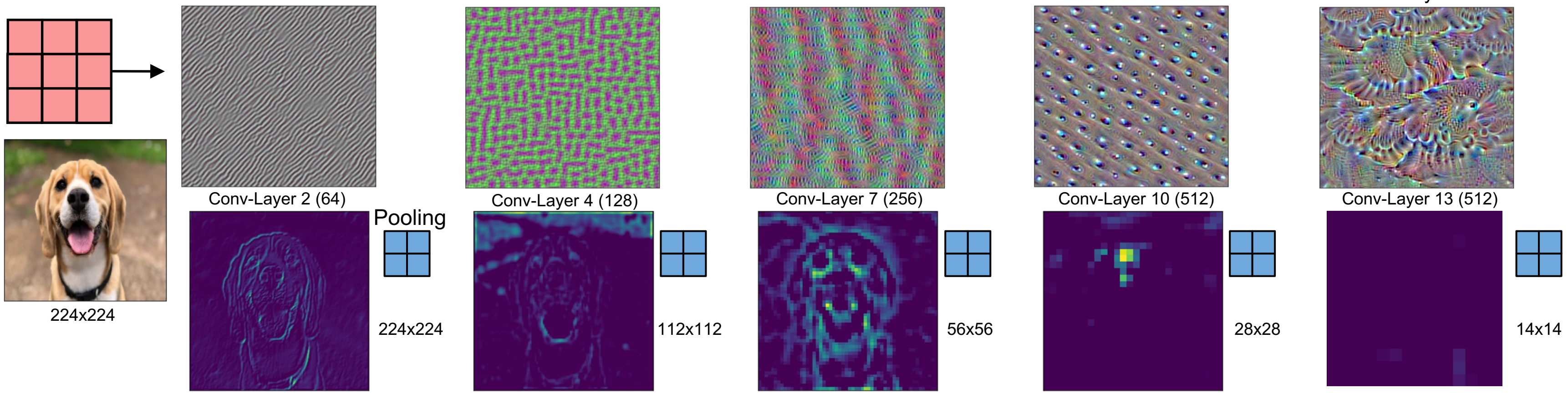
Related Work – Convolutional Layers + Pooling

- Convolutional layers: extraction of local features
- (Maximum) Pooling: reduction of resolution



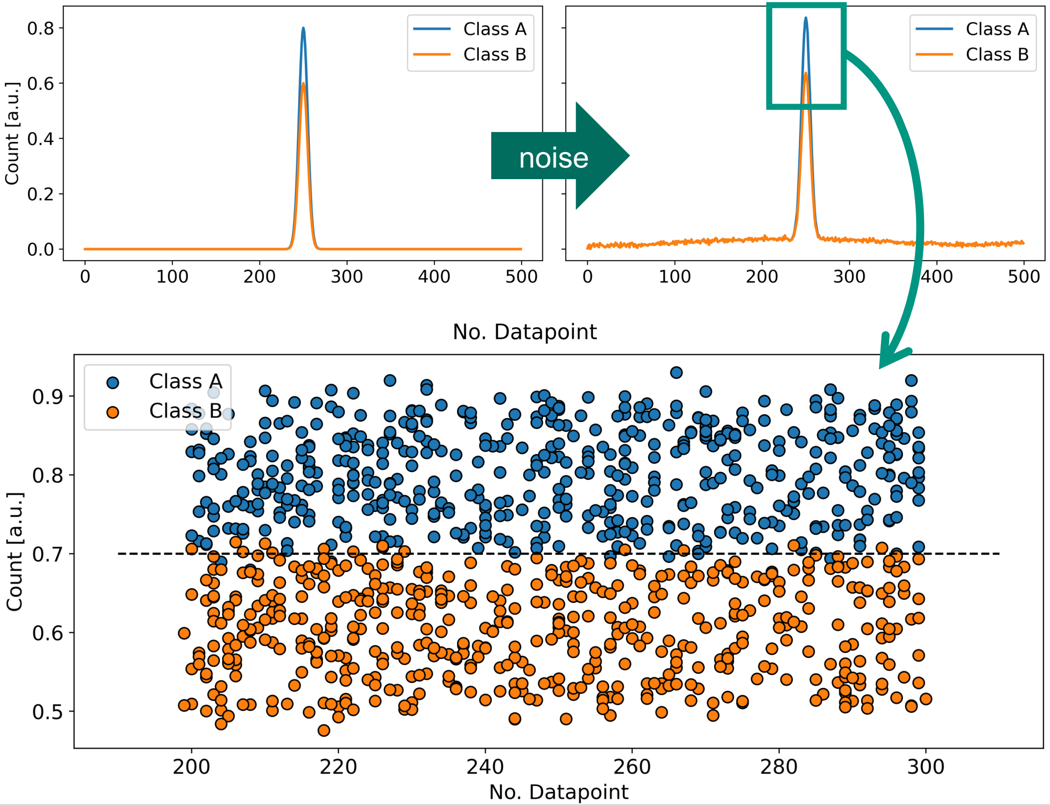
From Szymanski et al. 2021 [7]

VGG16 network, pretrained weights from Imagenet



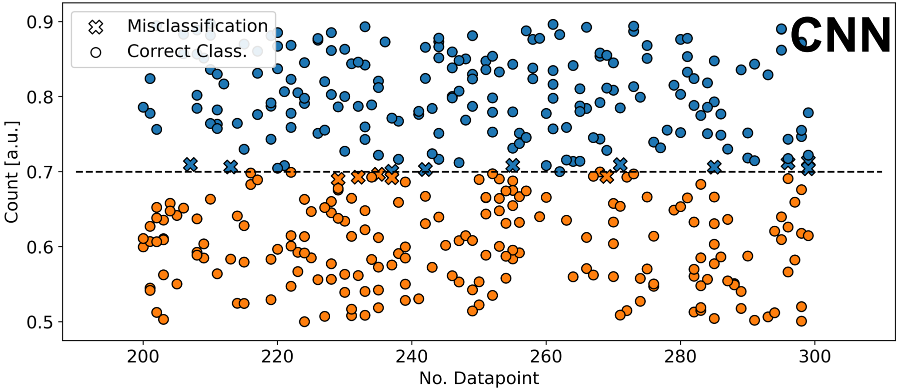
Evaluation Dataset – Training Samples

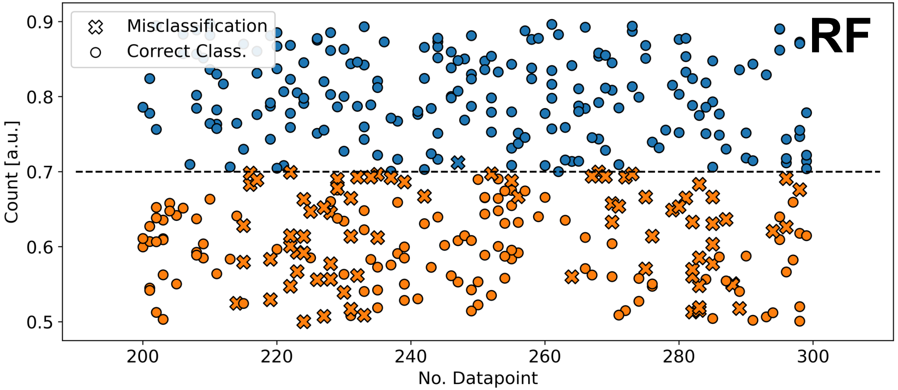
- Classification of single peak: max. intensity 0.8 or 0.6
- Variation of position (+/- 50), intensity (+/- 0.1) and shapes (Gaussians)
- Addition of background function and noise
- Result: minor overlap of max. intensities



Evaluation Dataset – Classification Results

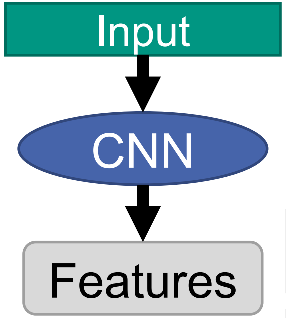
- Accuracy of CNN architecture [5] **96%**
- Performance of traditional ML: Random-Forest (RF) *80%*
- CNN distinguishes between both classes, while *RF performs worse*

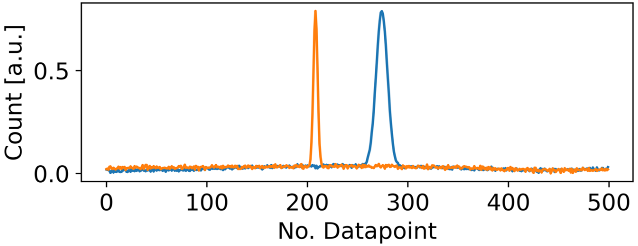


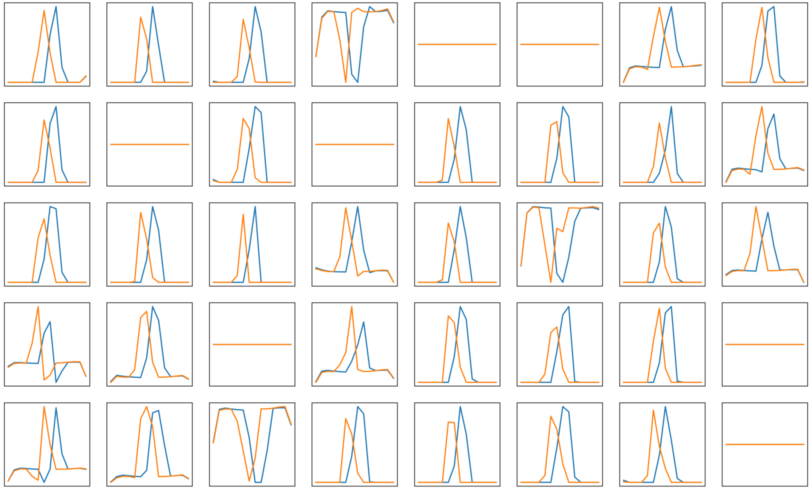


Evaluation Dataset – CNN Feature Maps

- What is the output of the convolutional layers?
- Reduction of
  - Noise
  - Background
  - Shape variation
  - Position variation



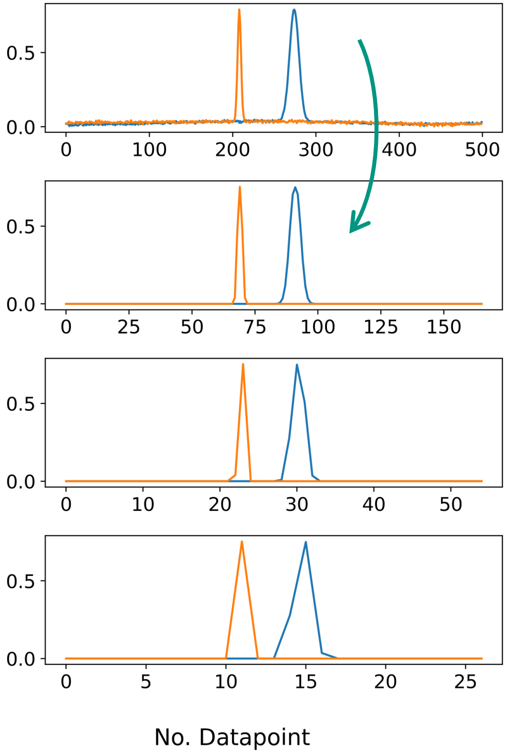




Evaluation Dataset – Benefits of MaxPooling

- Reduction of positional variation from MaxPooling
- How do traditional ML models benefit from reduced input?
- Second MaxPooling layer already improves performance from 92% to 94%
- <u>Similar performance of Random Forest *for reduced inputs*</u>

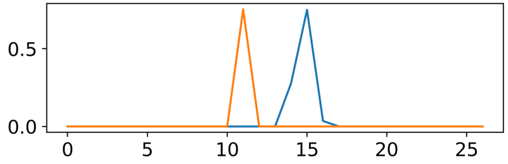
Input	Accuracy
Original	80%
After 1. MaxPool	81%
After 2. MaxPool	92%
After 3. MaxPool	93%



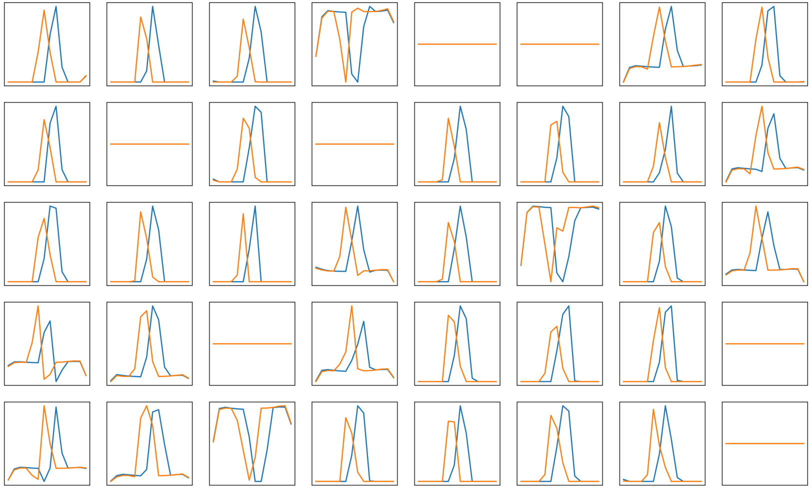
Evaluation Dataset – Contribution of Convolutions

- MaxPooling reduces position and shape variations
- What is the benefit of using Convolutional layers then?

➡ Conv-Layers eliminate background and match peak shapes to facilitate classification



vs.



Evaluation Dataset – Conv-Layer Configuration

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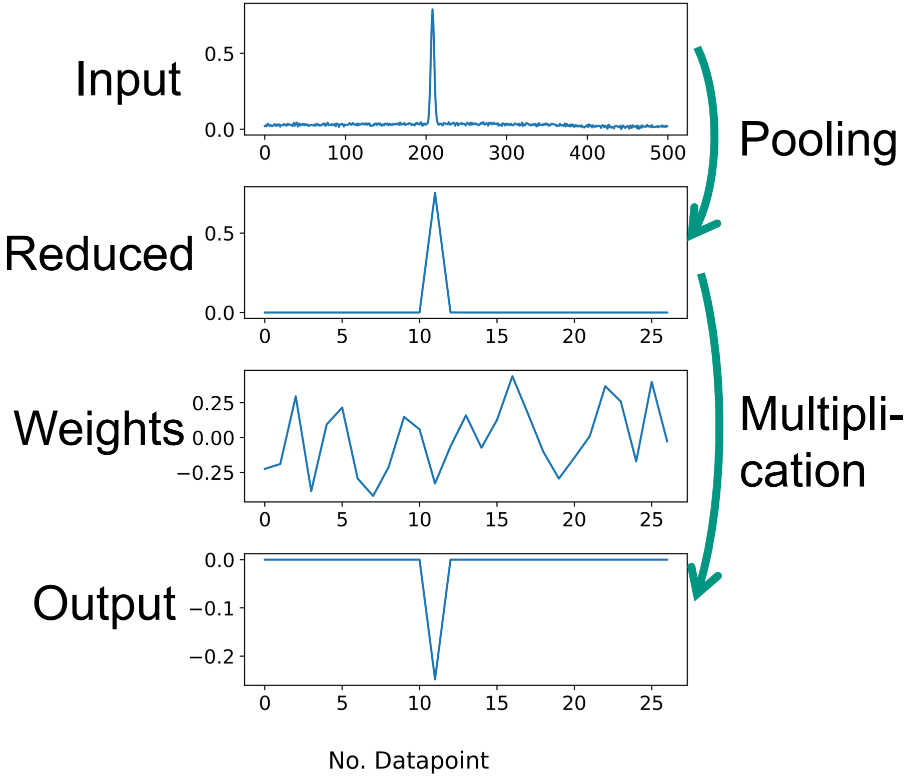
- CNN with single filter per layer? (reducing computational effort)
- Randomly initialized weights possibly cause negative peaks
- ReLU activation sets negative values to zero
  → output "empty"
- <u>Different initialization methods or activation function required</u>

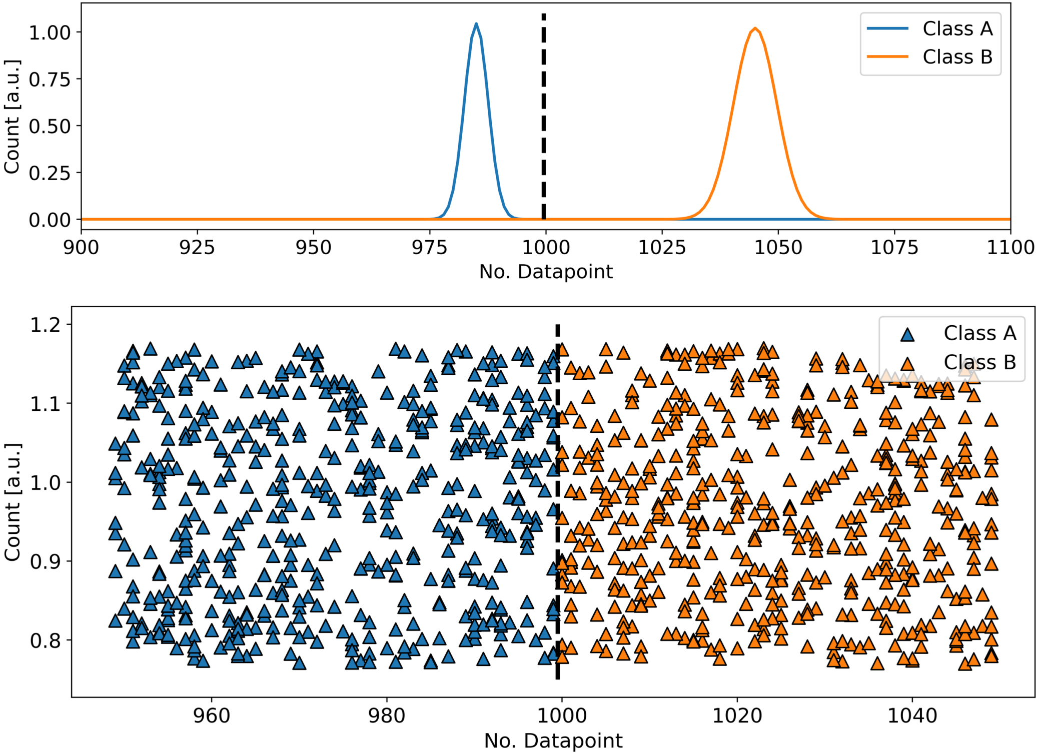


Recent Developments – Overview

- For images: CNNs with few convolutional layers state-of-the-art in 2012, advancement through stacking more convolutional layers and more complex structures (Resnet, Inception, etc.)
- For spectra: CNNs with 1-3 convolutional layers in 2017-2020, recently stacking more layers [7] or copying complex structures (Resnet) [8].
- More layers → Resolution of spectra gets even more reduced
- <u>BUT</u>: What if position of peaks is important for classification?

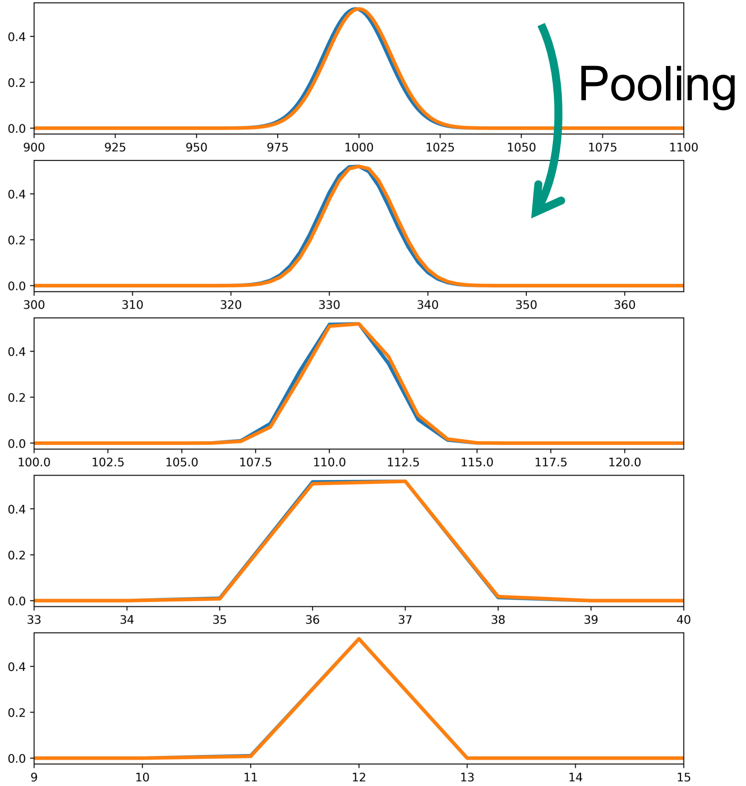
Recent Developments – Dataset

- Evaluating positional information with second dataset
- Class A: Max. at 950-999
  Class B: Max. at 1000-1050
  → No overlap
- Model: Resnet [8]



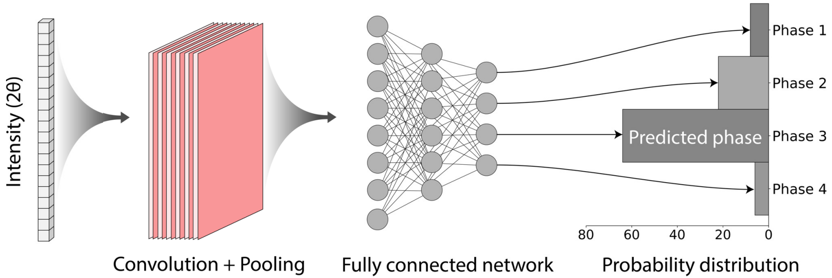
Recent Developments – Resnet Performance

- Resnet fails to correctly classify spectra with peak maxima close to border
- Pooling reduces resolution
  → **peaks align and become indistinguishable**
- Solution: Use *less* conv-layers/pooling



Related Work – Batch Normalization

- Batch Normalization as regularization
- Removing background + rescaling features
- Highlights “unique” features



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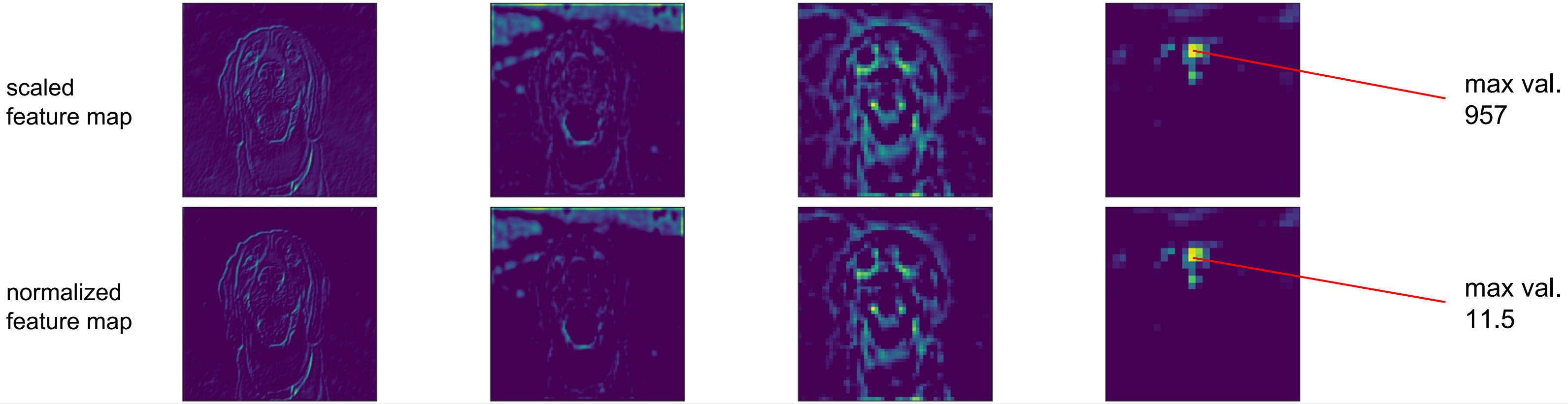
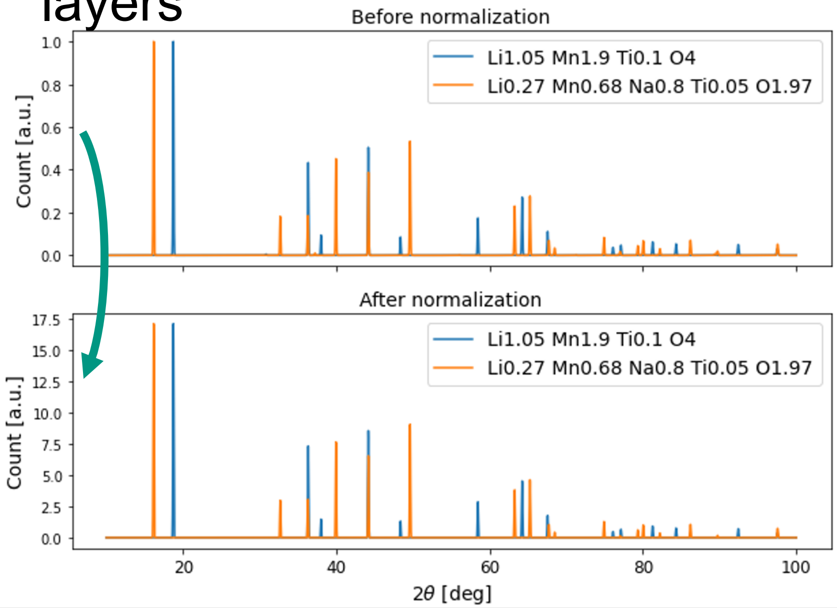


Recent Developments– Batch Norm. for spectra

- Recent networks like Resnet apply Batch-Norm. between convolutional layers



- No “unique“ features per class, nothing to *highlight*

- Normalization questionable for spectra

Conclusion

1. Convolutional layers work well on spectra because filters reduce peak shape variations + background and pooling reduces peak position shifts
2. Traditional ML algorithms struggle on peak shift variations but perform similarly as networks on lower resolution data
3. Spectra exhibit different ”features” compared to image data: adaptation of initialization or activation functions necessary
4. More elaborate structures & techniques developed for image data not better for spectra; always evaluate usage

References

[1] Schuetzke et al. “Enhancing deep-learning training for phase identification in powder X-ray diffractograms.” *IUCrJ* 8 (2021):408-420.

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[3] Liu et al. “Deep Convolutional Neural Networks for Raman Spectrum Recognition: A Unified Solution.” *The Analyst* 142 (2017): 4067-4074.

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[5] Lee et al. “A deep.-learning technique for phase identification in multiphase inorganic compounds using synthetic XRD powder patterns.” *Nature Communications* 11 (2020).

[6] Schuetzke et al. “A universal synthetic dataset for machine learning on spectroscopic data.” *Preprint on arXiv* (2022).

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