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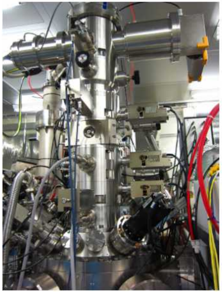

Machine Learning Enable Lineshape Analysis in Optical Two-Dimensional Coherent Spectroscopy

Michael Titze, Srikanth Namuduri, Shekhar Bhansali, Hebin Li

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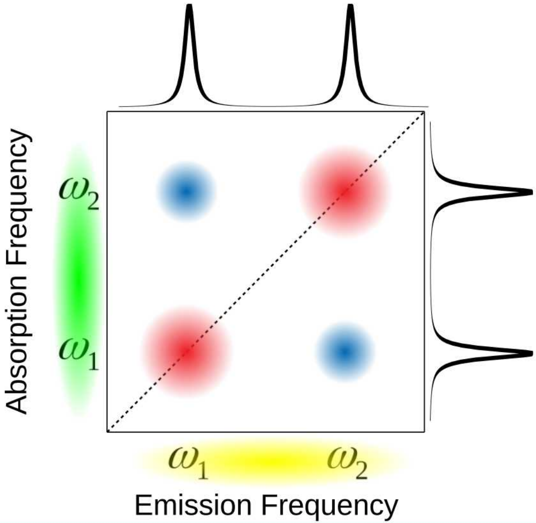
Advantages of Two-Dimensional Spectroscopy

Two-Dimensional Spectroscopy has two main advantages over non-2D spectroscopies

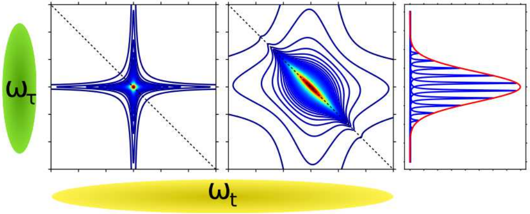
1. Deconvolution of congested and complex spectra along two dimensions



Advantages of Two-Dimensional Spectroscopy

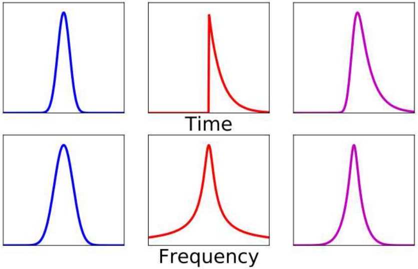
Two-Dimensional Spectroscopy has two main advantages over non-2D spectroscopies

1. Deconvolution of congested and complex spectra along two dimensions
2. Simultaneous detection of homogeneous and inhomogeneous linewidths



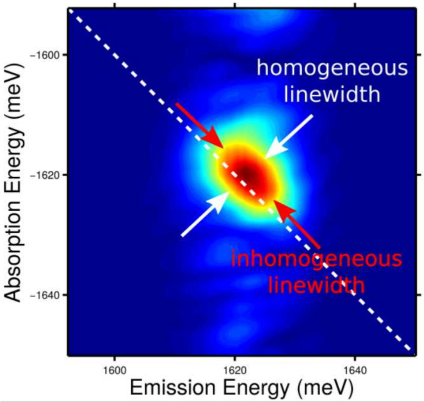
Linewidth Measurement using 2D Spectroscopy Data

- Linewidth measurements are necessary for quantitative analysis of spectra
- Qualitative analysis can determine if spectrum is homogeneously or inhomogeneously broadened
- In case of similar homogeneous and inhomogeneous broadening, homogeneous and inhomogeneous linewidths are entangled



Linewidth Measurement using 2D Spectroscopy Data

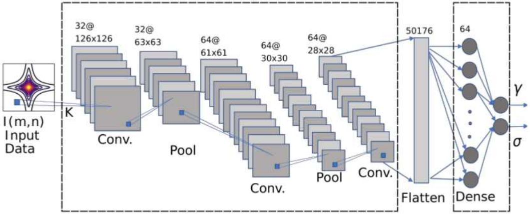
- Linewidth measurements are necessary for quantitative analysis of spectra
- Qualitative analysis can determine if spectrum is homogeneously or inhomogeneously broadened
- In case of similar homogeneous and inhomogeneous broadening, homogeneous and inhomogeneous linewidths are entangled
- Analytic expressions were derived by Siemens et al. for short pulses and Smallwood et al. for finite pulses



M. Titze et al., Phys. Rev. Materials 2, 054001 (2018)

Machine Learning for Analysis of 2D Spectra

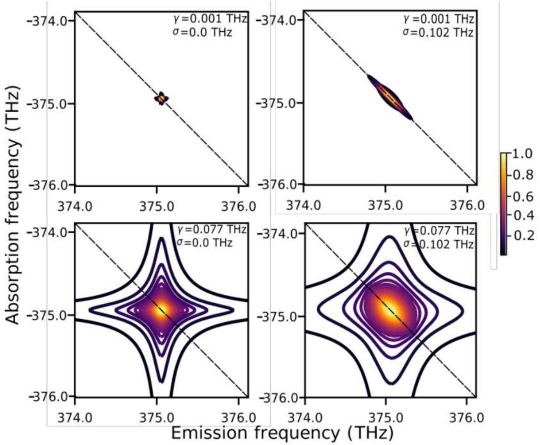
- Two main issues with manual fitting of 2D spectra
  1. Operator time: Multi-parameter nonlinear fitting requires tedious adjustment of initial values
  2. Computation time: Algorithm training only needs to be done once, while fitting routine needs to be run for each spectrum
- Here use convolutional neural network based machine learning algorithm
  - Fully connected network in principle better, but much more computation intense



S. Namuduri et al., JOSA B ????

Training the Machine Learning Algorithm

- To save experimental time, use simulated data to train the network, total 4096 spectra are simulated
  - 3686 spectra are used to train the network
  - 410 spectra are set aside for later testing of the trained network



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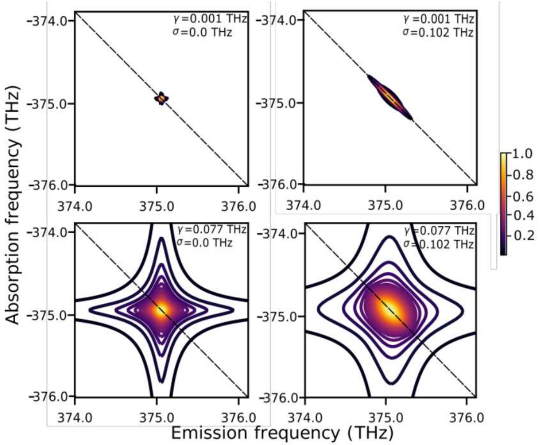
Training the Machine Learning Algorithm

- To save experimental time, use simulated data to train the network, total 4096 spectra are simulated
- To train the network, need a loss function
  - Here use RMS error of homogeneous and inhomogeneous linewidth
  - NN is adjusting weights of each node by minimizing the loss function



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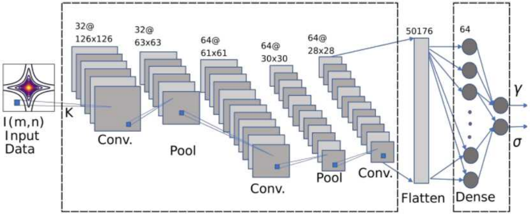
Network Architecture

- CNNs have been found to be well-suited for analysis of image data
- 2D Spectroscopy data is a 2D map, can be considered image data
- Our network consists of
  1. 3 convolutional layers
  2. 2 pooling layers
  3. 1 flatten layer
  4. 1 fully-connected dense layer
  5. 1 output layer
- To evaluate the CNN, we use RMS error and percent error on the output results



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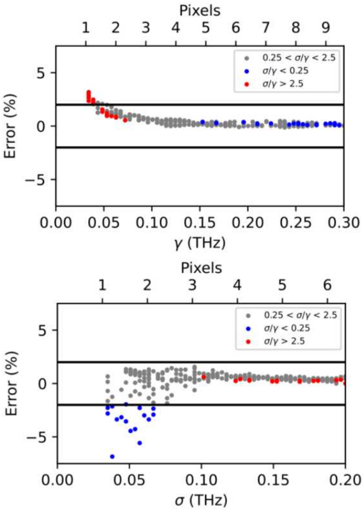
Testing the Machine Learning Algorithm

- Use the 410 spectra that were put aside to determine homogeneous and inhomogeneous linewidths, then compare to actual linewidth
- Algorithm does well for intermediate broadening, when either homogeneous or inhomogeneous width get too small/large, accuracy is lower



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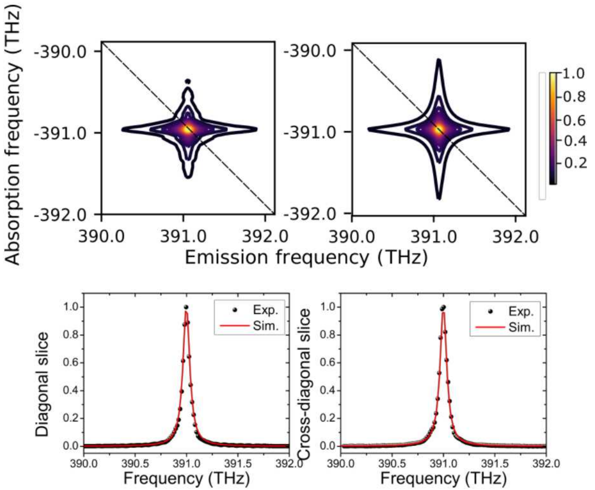
Testing Against Real Data: Homogeneously Broadened

- Use K-Vapor D-lines, homogeneously broadened system
- Measure four peaks, only investigate D1-line
- Use parameters found by ML algorithm to simulate a spectrum
- Compare overall shape and linecuts



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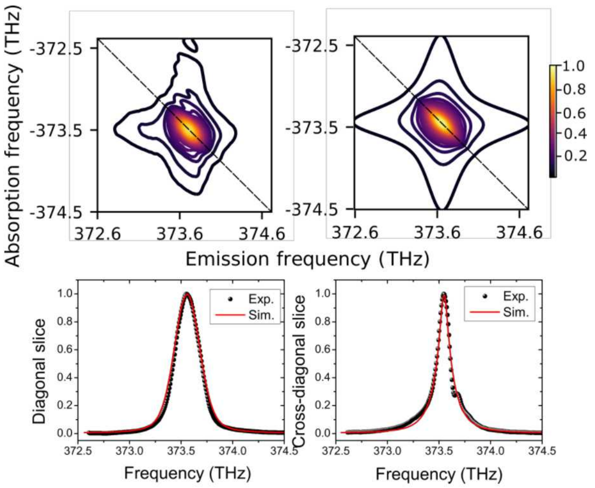
Testing Against Real Data: Inhomogeneously Broadened

- GaAs Quantum Well, intermediate inhomogeneously broadend system
- Use parameters found by ML algorithm to simulate a spectrum
- Compare overall shape and linecuts



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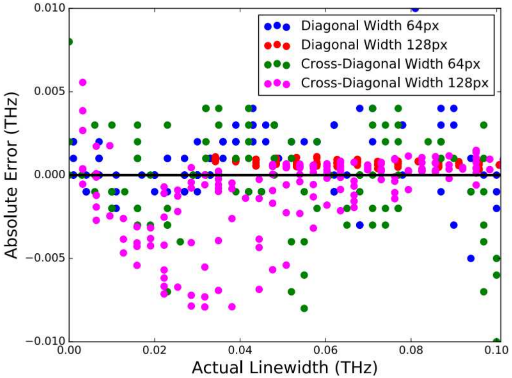
Evaluating the ML Algorithm

- Improvement with higher pixel number on training data

Array Size	Samples	Avg Error
32×32	1024	$> 10\%$
32×32	2048	8.5%
64×64	1024	6%
128×128	1024	4%
128×128	4096	$< 2\%$

- ML algorithm drastically reduces machine time, 3200 ms using manual fit routine vs 80 ms using ML algorithm



Conclusion

- High-speed algorithm for extracting linewidths from 2D spectroscopy data
- No operator input required, ideal for on-the-fly data analysis
- Good accuracy for intermediately broadened case, hard case for manual fitting
- Demonstrated capability of accurately measuring noisy real experimental data