Physikalisch-Technische Bundesanstalt
Braunschweig and Berlin
National Metrology Institute

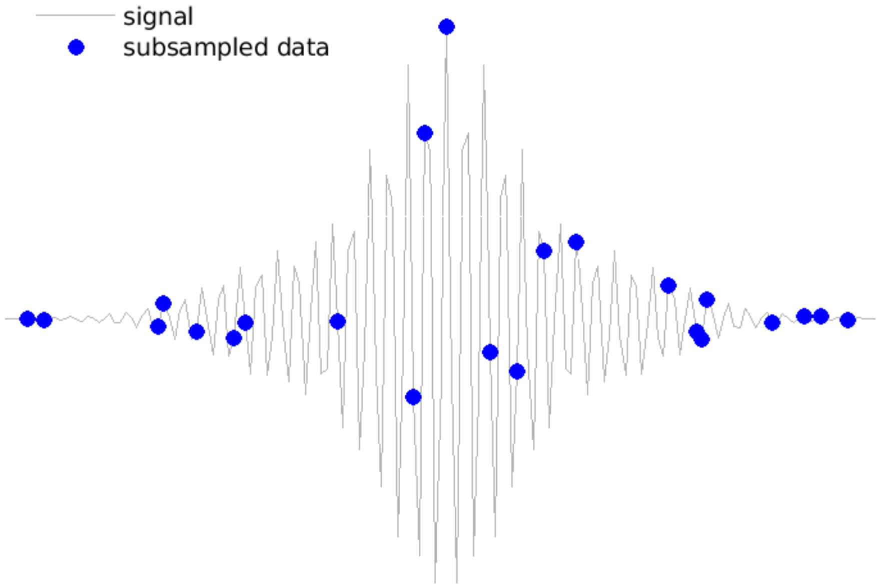
Joint regression and compressed sensing for chemical mapping in nano-FTIR

G. Wübbeler, M. Marschall, E. Rühl, B. Kästner, & C. Elster

Content

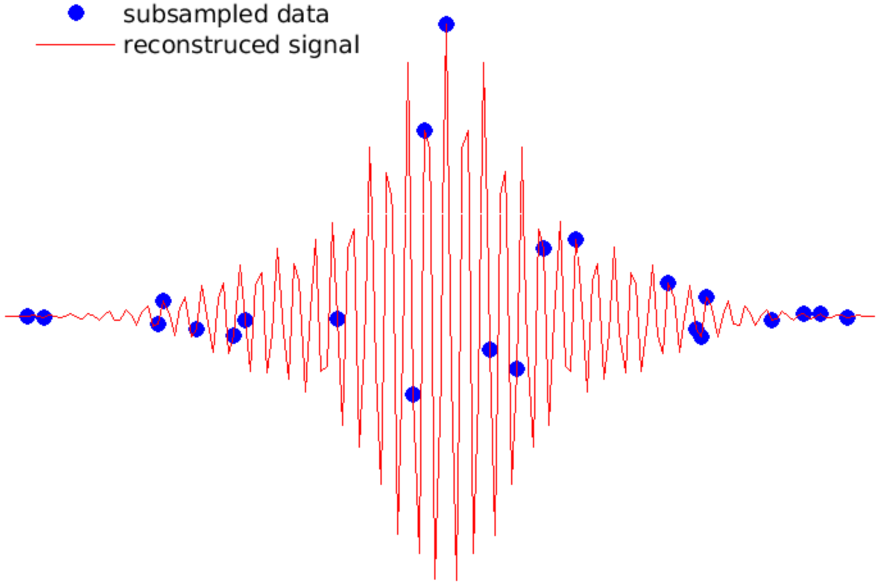
- **Compressed sensing**
- **Joint regression and compressed sensing**
- **Application to Nano-FTIR**
- **Summary**

Compressed sensing



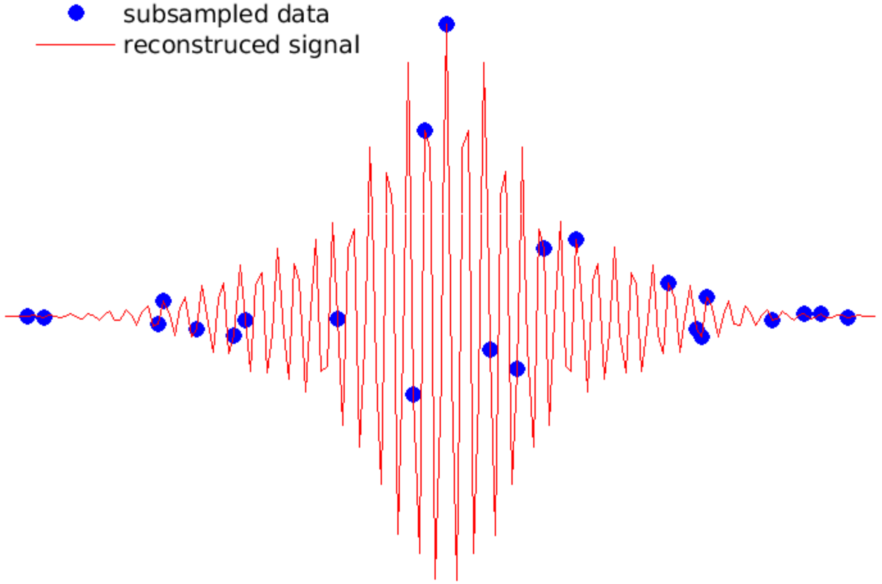
Goal: signal reconstruction given randomly subsampled data

Compressed sensing



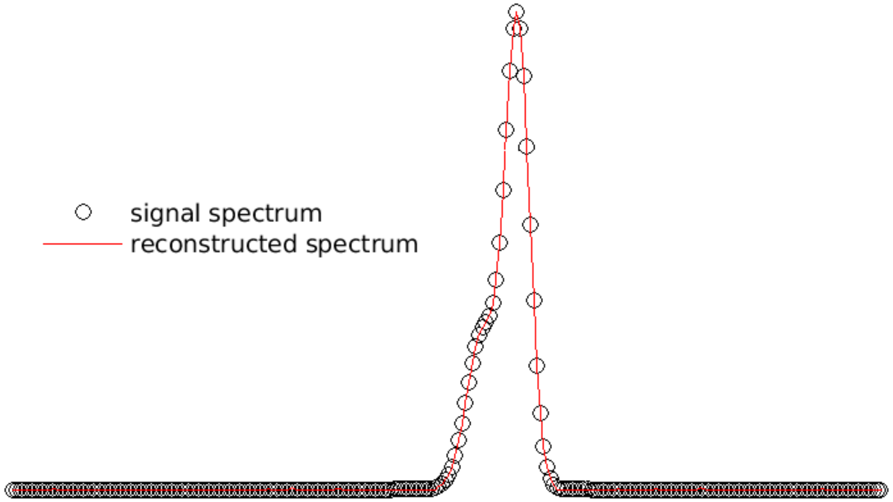
Compressed sensing reconstruction

Compressed sensing



Compressed sensing reconstruction

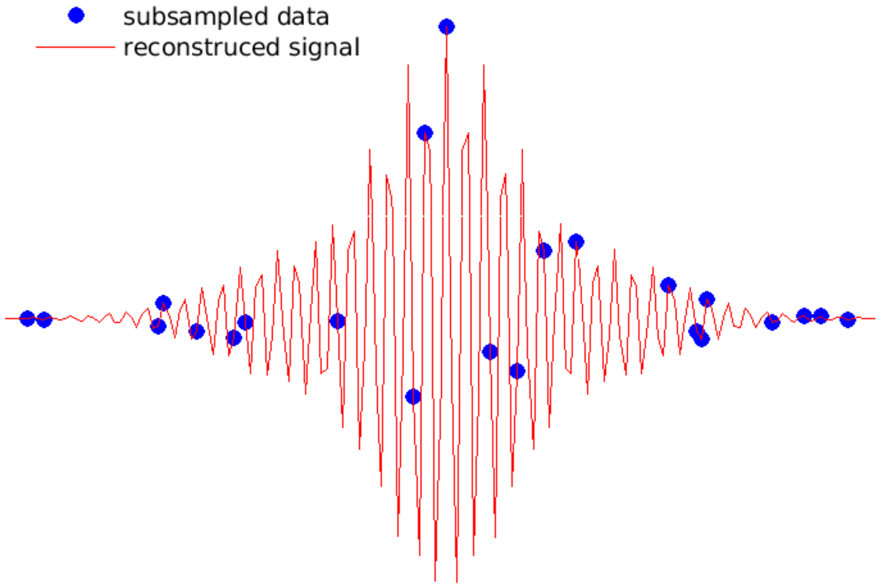
Fourier domain

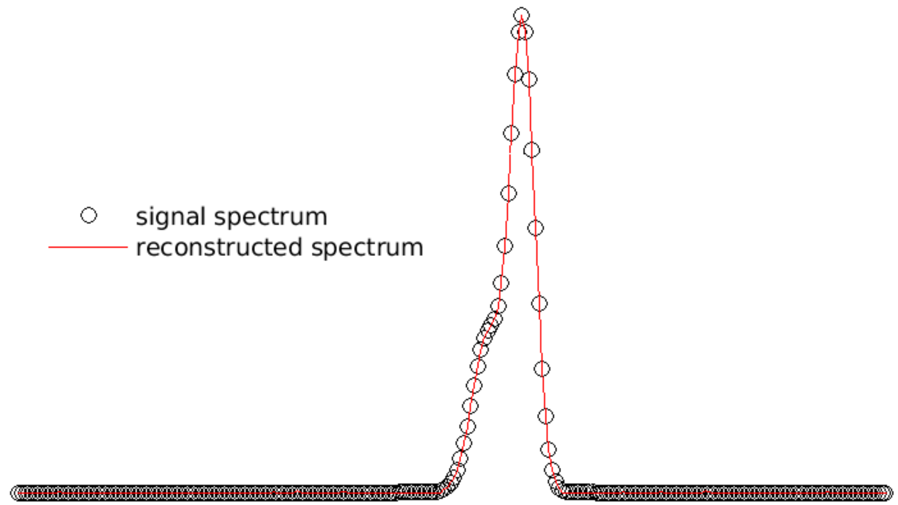


Only few non-zero Fourier coefficients

Compressed sensing

Fourier domain





Compressed sensing reconstruction

Only few non-zero Fourier coefficients

Signal is sparse in Fourier domain

Compressed sensing: Math

Sparse recovery from an underdetermined linear system

$$\boxed{\min_{\mu} \|\mu\|_0 \quad \text{subject to} \quad y = V\mu}$$

y	subsampled data
V	known sensing matrix
μ	sought sparse vector
$\|\mu\|_0$	number of non-zero elements

Compressed sensing: Math

Sparse recovery from an underdetermined linear system

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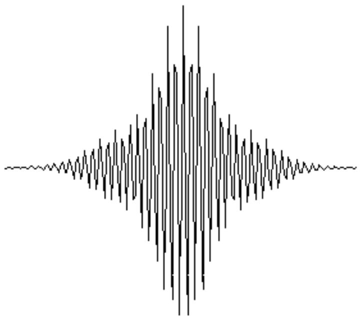
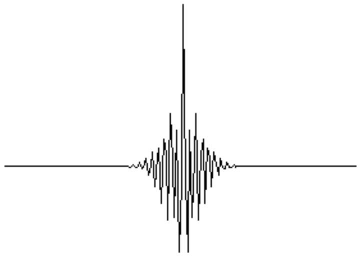
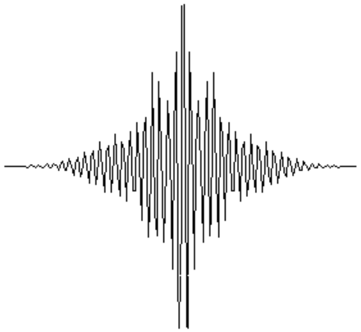
Solution via combinatorial search usually not tractable

- Convex relaxation : $\min_{\mu} \|\mu\|_1$ subject to $y = V\mu$

- Alternative: Greedy methods

Donoho, D. L. (2006). Compressed sensing. *IEEE Transactions on information theory*, *52*(4), 1289-1306.
Candes, E. J. (2008). The restricted isometry property and its implications for compressed sensing. *Comptes rendus mathematique*, *346*(9-10), 589-592.
Eldar, Y. C., & Kutyniok, G. (Eds.). (2012). Compressed sensing: theory and applications. Cambridge University Press.

Joint regression and compressed sensing

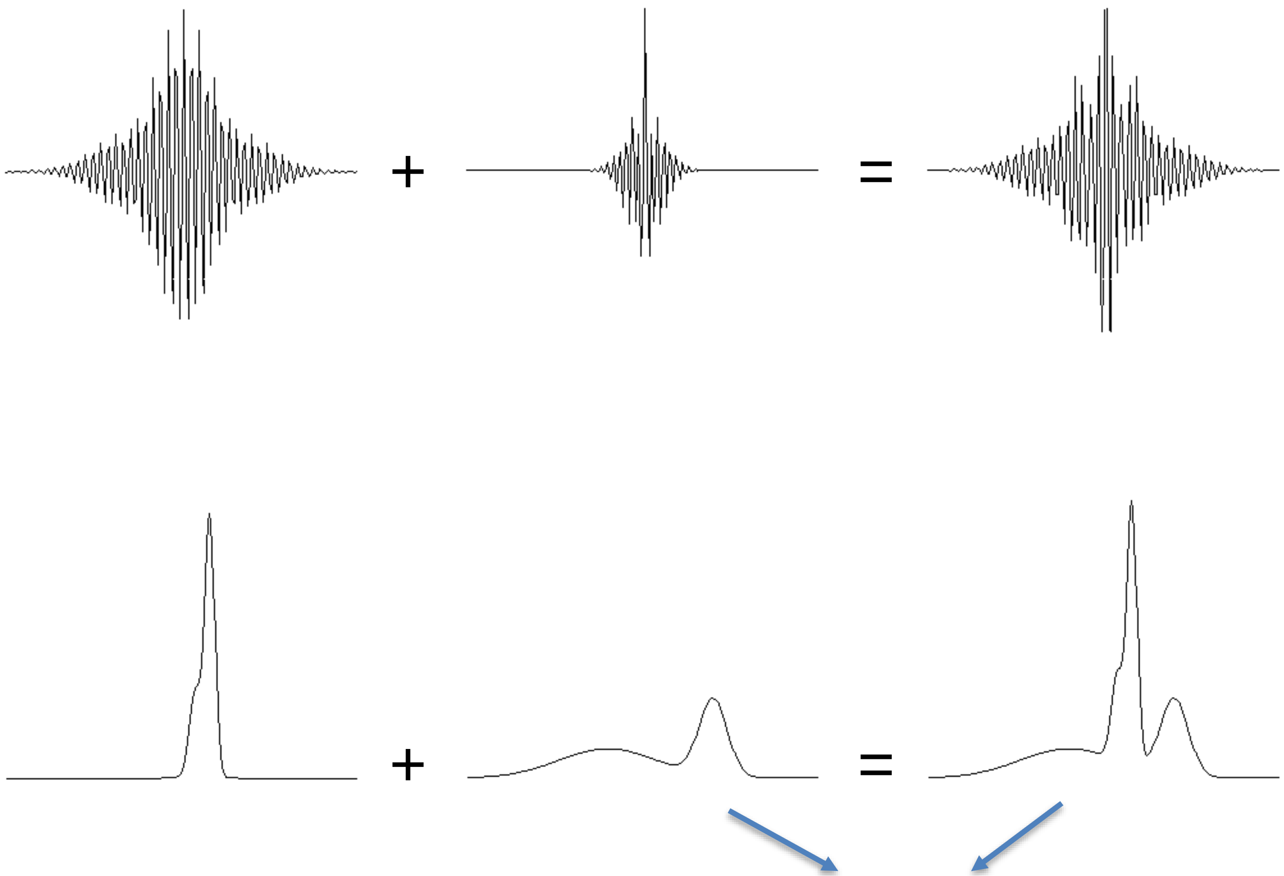
	sparse component		regression component		
signal domain		+		=	

Joint regression and compressed sensing

sparse component **regression component**

signal domain

Fourier domain



possibly non-sparse

Joint regression and compressed sensing

Model $$y = V\mu + X\theta$$

sparse component **regression component**

y	subsampled data
V	known sensing matrix
μ	sought sparse vector
X	known design matrix
θ	sought weights (few)

Joint regression and compressed sensing

Model $$y = V\mu + X\theta$$

$\downarrow$ **sparse component** $\searrow$ **regression component**

y	subsampled data
V	known sensing matrix
μ	sought sparse vector
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θ	sought weights (few)

Two step procedure

1) Evaluation of sparse signal

- Determine matrix P so that $PX = 0$
- $\tilde{y} = Py = PV\mu$ represents standard compressed sensing task
- Apply greedy method to obtain sparse representation

Joint regression and compressed sensing

Model $y = V\mu + X\theta$

sparse component **regression component**

y	subsampled data
V	known sensing matrix
μ	sought sparse vector
X	known design matrix
θ	sought weights (few)

Two step procedure

2) Regression

- Reduced regression task $y = \tilde{V}\,\tilde{\mu} + X\theta$

  $\tilde{V}$: matrix containing only columns of V where $\mu \neq 0$

- Identifiable since $\mathrm{rank}([\tilde{V}, X]) = p + r$

  p number of weights θ

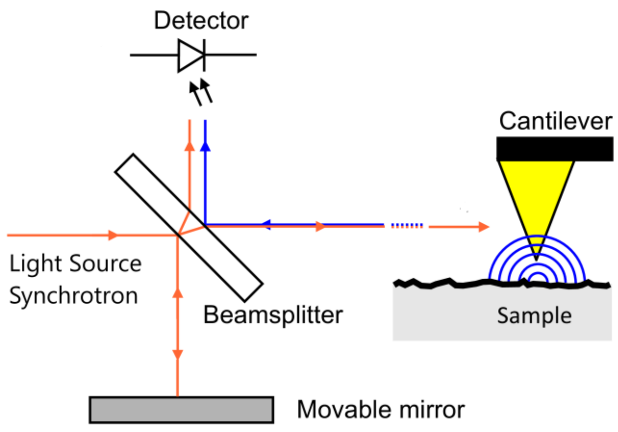
  r number non-zero elements of μ

➔ Estimated weights θ and coefficients $\tilde{\mu}$

Nano-Fourier-transform infrared spectroscopy (nano-FTIR)

- Combines infrared (IR) spectroscopy with scanning probe microscopy.

- Enables hyperspectral imaging at nanometer spatial resolution



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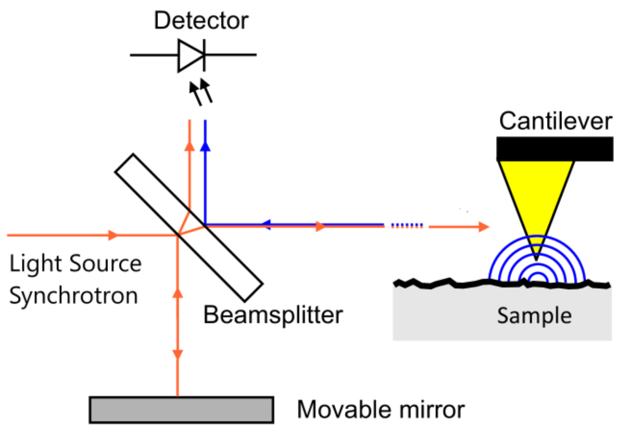

Nano-FTIR can be used to determine **chemical mappings**

- Spatial distribution of concentration of substances contained in the sample
- Obtained by solving a regression task using known spectral characteristics

Huth F, Govyadinov A, Amarie S, Nuansing W, Keilmann F and Hillenbrand R (2012) Nano-FTIR absorption spectroscopy of molecular fingerprints at 20 nm spatial resolution *Nano Lett.* **12** 3973–8

Amarie S, Zaslansky P, Kajihara Y, Griesshaber E, Schmahl W W and Keilmann F 2012 Nano-FTIR chemical mapping of minerals in biological materials *Beilstein J. Nanotechnol*. 3 312–23

Nano FTIR chemical mapping

Challenges

- Nano-FTIR is scanning based ➔ long measurement times (hours)
- Data may contain signal contributions of further, unknown substances

Nano FTIR chemical mapping

Challenges

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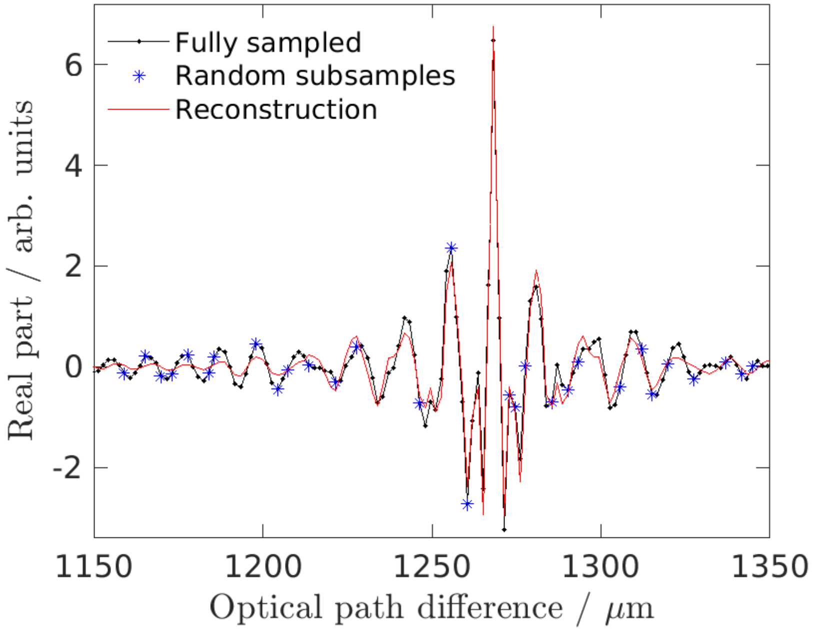
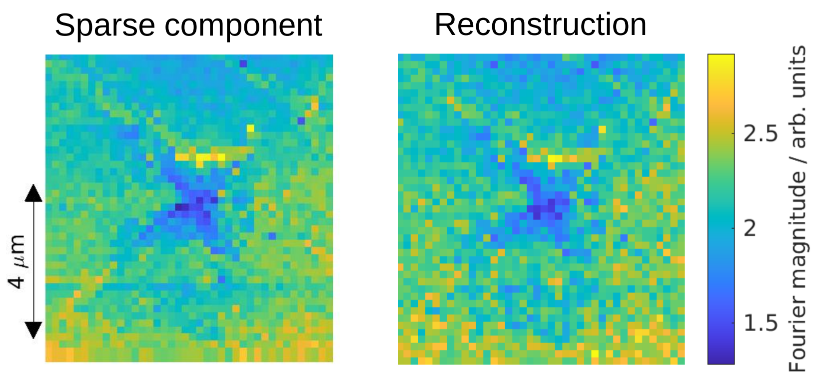
Joint regression and compressed sensing approach

- Subsampling enables reduced measurement times
- Additional signal contribution is assumed to be sparse
- Spatial regression includes Gaussian Markov random field regularization
  Rue, H., & Held, L. (2005). Gaussian Markov random fields: theory and applications. Chapman and Hall/CRC.

Nano FTIR reconstruction

- Real nano-FTIR measurements*) superimposed by simulated chemical mappings
- Simulated components and sparse signal exhibit spectral overlap
- Data taken at a subsampling rate of 20%.





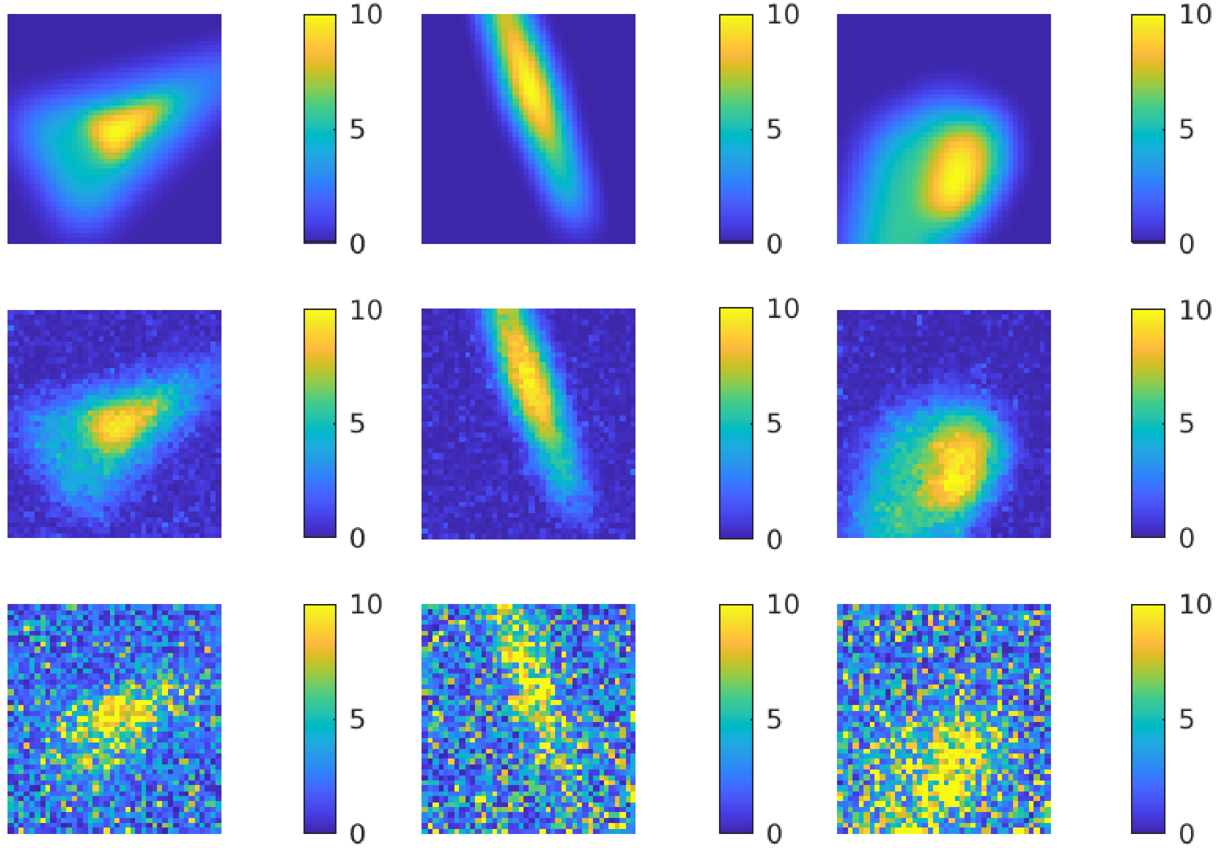
*) Kästner, B., Schmähling, F., Hornemann, A., Ulrich, G., Hoehl, A., Kruskopf, M., Pierz, K., Raschke, M. B., Wübbeler, G. and Elster, C. (2018) Compressed sensing FTIR nano-spectroscopy and nanoimaging. Optics Express, vol. 26, no. 14, pp. 18115-18124.

Chemical mapping estimates

Simulated mappings

Joint regression and compressed sensing

Naïve regression, sparse signal treated as error



Summary

- Development of a joint regression and compressed sensing approach
- Subsampling enables reduced measurement times
- Unknown signal contributions modelled non-parametrically
- Functionality demonstrated using augmented nano-FTIR data

Wübbeler, G., Marschall, M., Rühl, E., Kästner, B., & Elster, C. (2022). Compressive nano-FTIR chemical mapping. Measurement Science and Technology, 33(3), 035402.