Joint ESS ILL User Meeting

Autonomous Experiments
in
Inelastic Neutron Scattering

M. Boehm
Institut Laue-Langevin

5 October 2022

Neutron spectroscopy @ ILL:

https://www.ill.eu/users/scientific-groups/spectroscopy/

Spin-Echo:

WASP, IN15

Neutron back-scattering:

_IN13_* _(CRG CNR/UJA)_, **IN16b**

Time-of-flight:

Panther, IN5, _Sharp+_* (_CRG LLB_)

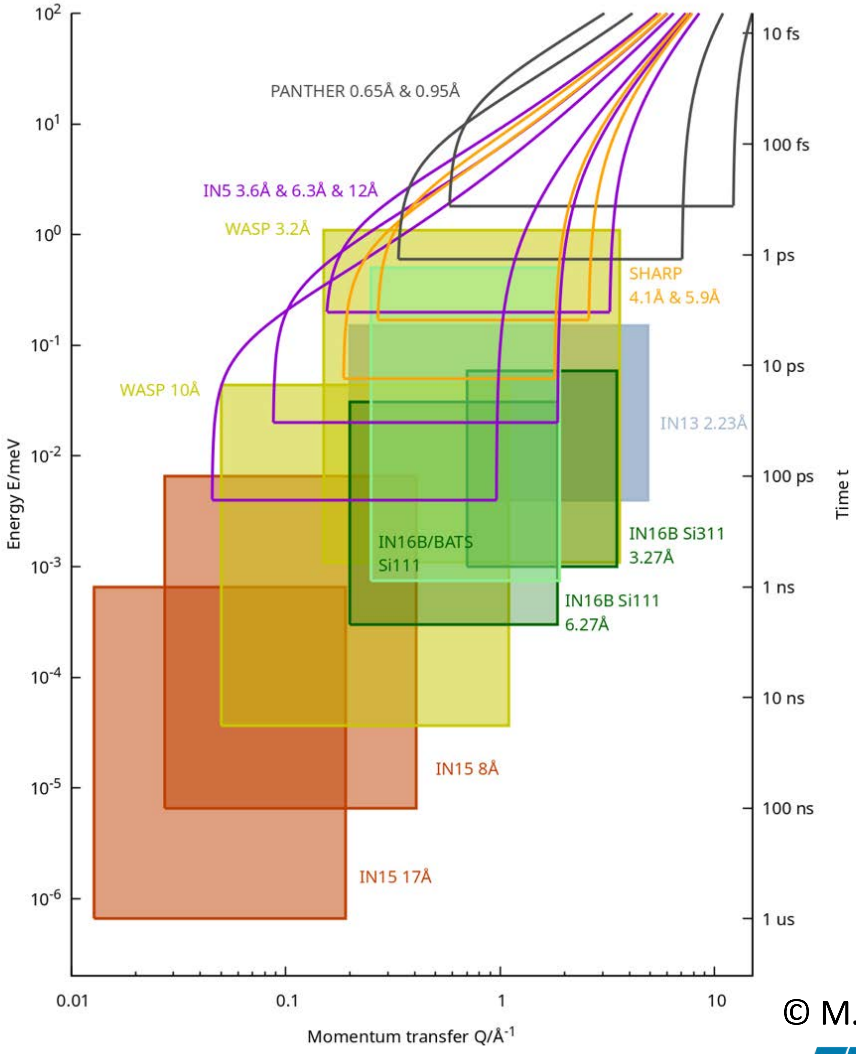
Three-Axis:

IN8, IN12* _(CRG JNCS/CEA)_, **ThALES, IN20, IN22*** _(CRG CEA/JCNS)_

Single x-stal spectroscopy

Vibrational:

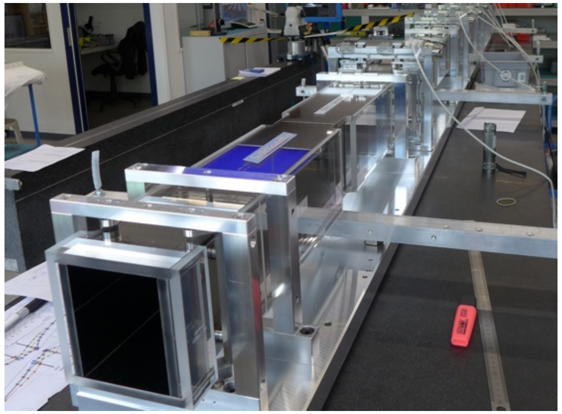
Lagrange



© M. Appel

WASP	New Fresnel: doubling of Ft (→15 ns @ 7Å). New vacuum box Extension analyzer banks from 90° to 150°
IN15	Fresnel upgrade (Trans x 2, Bg / 2). 5 position sample changer with in-situ DLS.
IN13* *(CRG CNR/UJA)*	New primary spectrometer (guide, mono). Work on detector upgrade
IN16b	New motorized focusing guide. BATS (TOF option) chopper upgrade
Panther	Fully integrated into user program. Installation of Bg choppers. PASTIS-3 in progress.
IN5	
Sharp+* *(CRG LLB)*	Commissioning of primary spectrometer in 2024
IN8	4 new monochromators, Thermes secondary spectrometer commissioned
IN12* *(CRG JNCS/CEA)*	
IN20	New PG002 mono and ana. New Heusler in 2024. NVS commissioned. PASTIS-3 in user program.
IN22* *(CRG CEA/JCNS)*	Extension experimental zone.
ThALES	New detector shielding
Lagrange	New low T sample changer. Work on in-situ Raman option

Fresnel coil @ WASP



Focusing guide @ IN16b

https://www.ill.eu/users/scientific-groups/spectroscopy/

Single crystal spectroscopy:

nature COMMUNICATIONS

ARTICLE

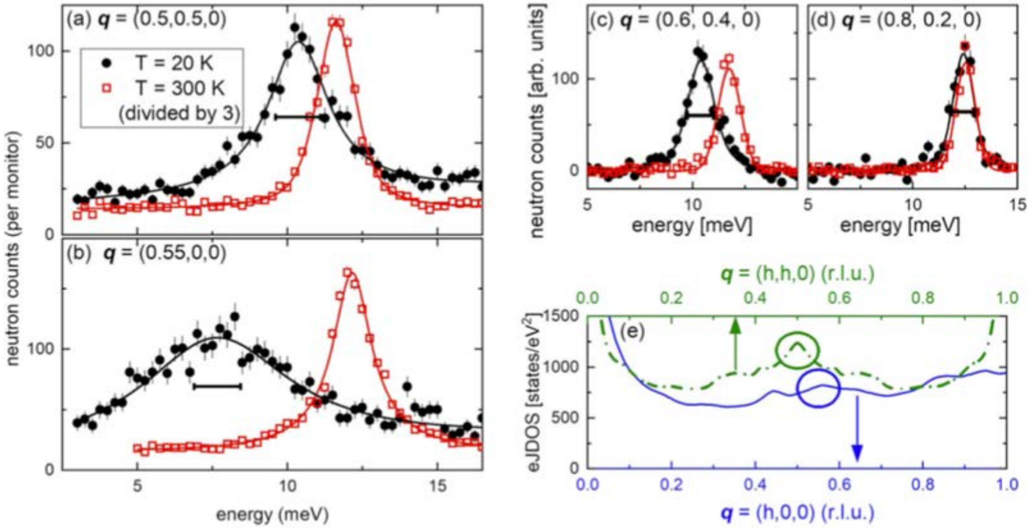
Check for updates

https://doi.org/10.1038/s41467-021-27843-y OPEN

Electron-momentum dependence of electron-phonon coupling underlies dramatic phonon renormalization in YNi_2B_2C

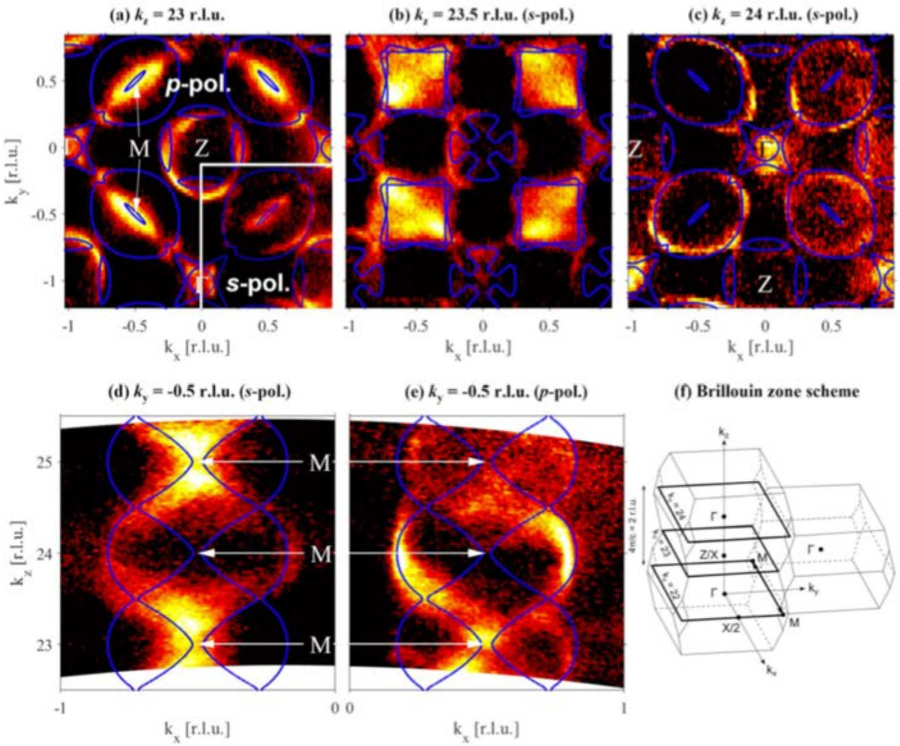
Philipp Kurzhals[1], Geoffroy Kremer[2], Thomas Jaouen[2,3], Christopher W. Nicholson[2], Rolf Heid[1], Peter Nagel[1], John-Paul Castellan[1,4], Alexandre Ivanov[5], Matthias Muntwiler[6], Maxime Rumo[2], Bjoern Salzmann[2], Vladimir N. Strocov[6], Dmitry Reznik[7,8], Claude Monney[2] & Frank Weber[1]✉

INS @ IN8:



P. Kurzhals et al., Nat Commun **13**, 228 (2022).

SX-ARPES @ ADRESS (SLS)



SC induced phonon anomalies

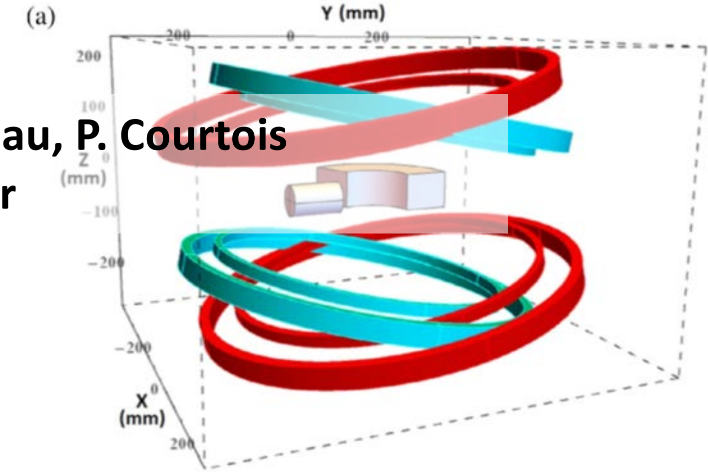
Combined soft x-ray ARPES and INS

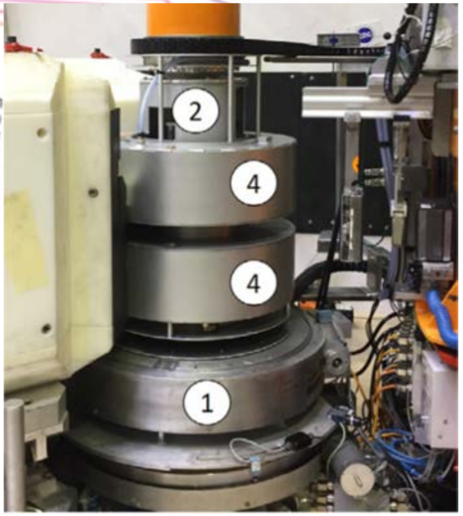
Ab-initio lattice and electronic dynamical calculations

XYZ wide angle PA on IN20 and Panther PASTIS-3:

D. Jullien, A. Petoukhov, N. Thiery, P. Mouveau, P. Courtois
U.B. Hansen, M. Enderle, B. Fåk, P. Chevallier

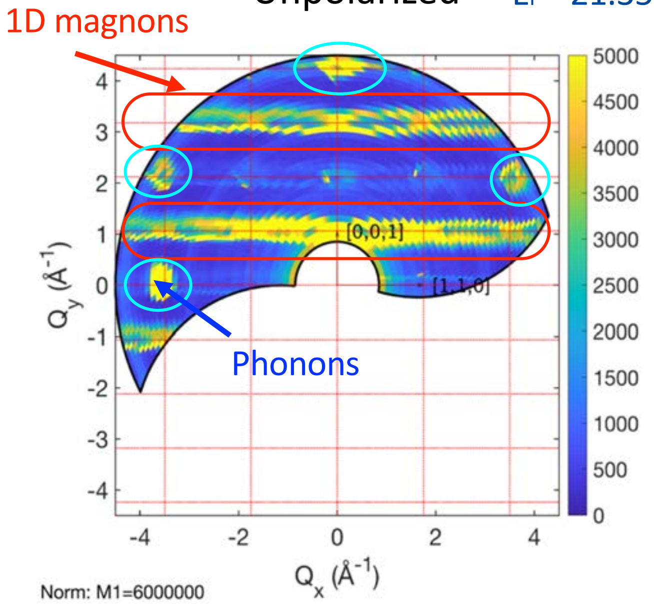
on IN20 available from next cycles
project on Panther in progress, ~ 2024



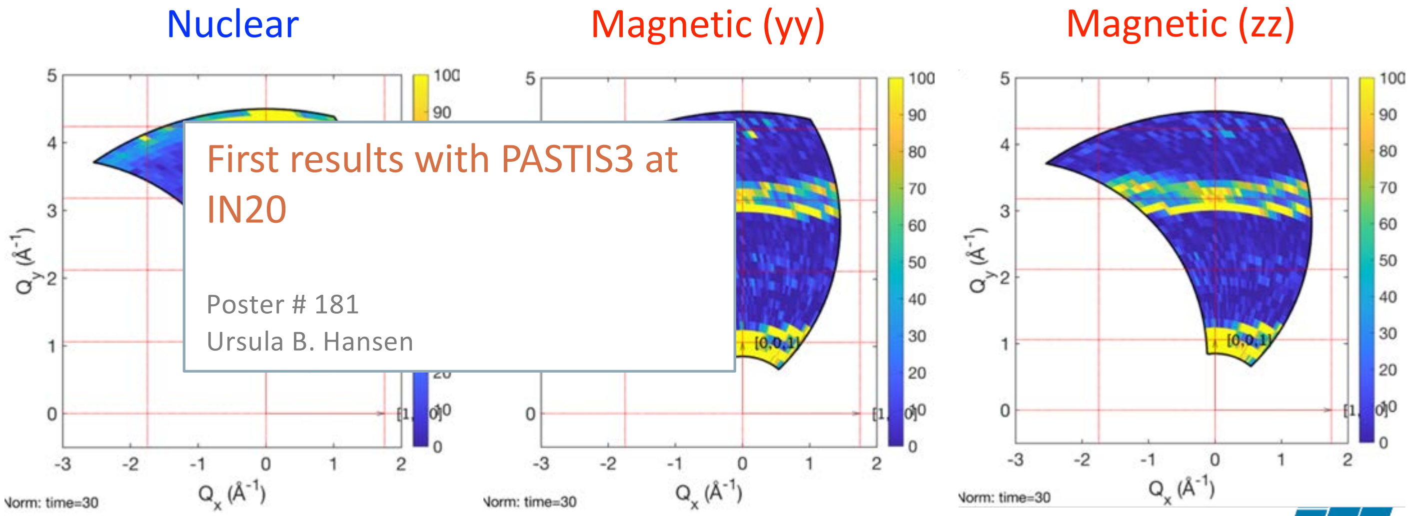


D. Jullien et al., Nucl.Instr & Meth in Phys.Res., A **1010** (2021), p. 165558

Unpolarized E_i = 21.35 meV, $\hbar\omega$ = 3 meV



Thanks to Ursula Hansen



First results with PASTIS3 at IN20

Poster # 181
Ursula B. Hansen

Single crystal spectroscopy - Exploring $S(\mathbf{Q},\omega)$:

Tendencies:

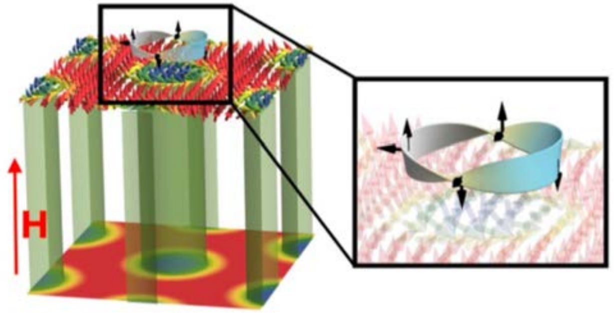
- Larger data volumes
- Complementarity
- Modelling/calculations
- New experimental possibilities

$\sum$ = beautiful science

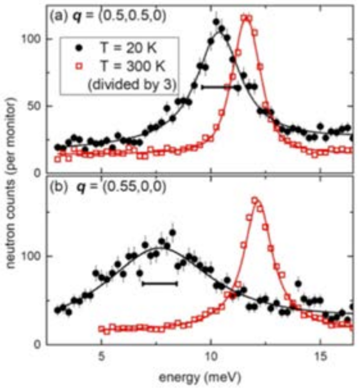
Consequences:

- Complexity in experiments
- Complex data analysis

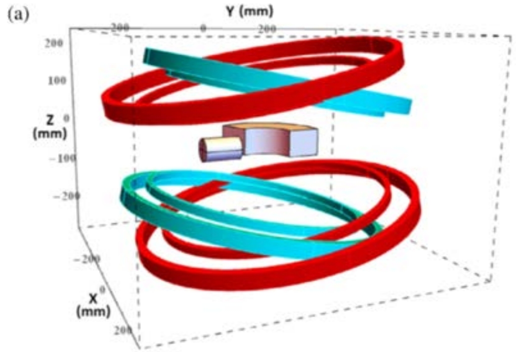
$\sum$ = publication rate?



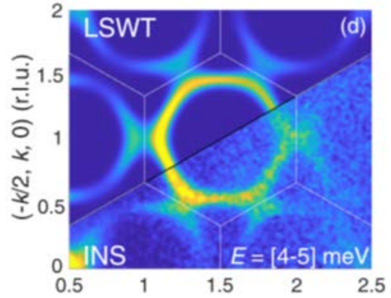
T. Weber et al., Science **375** (2022), p.1025



P. Kurzhals et al., Nat Commun **13**, 228 (2022).

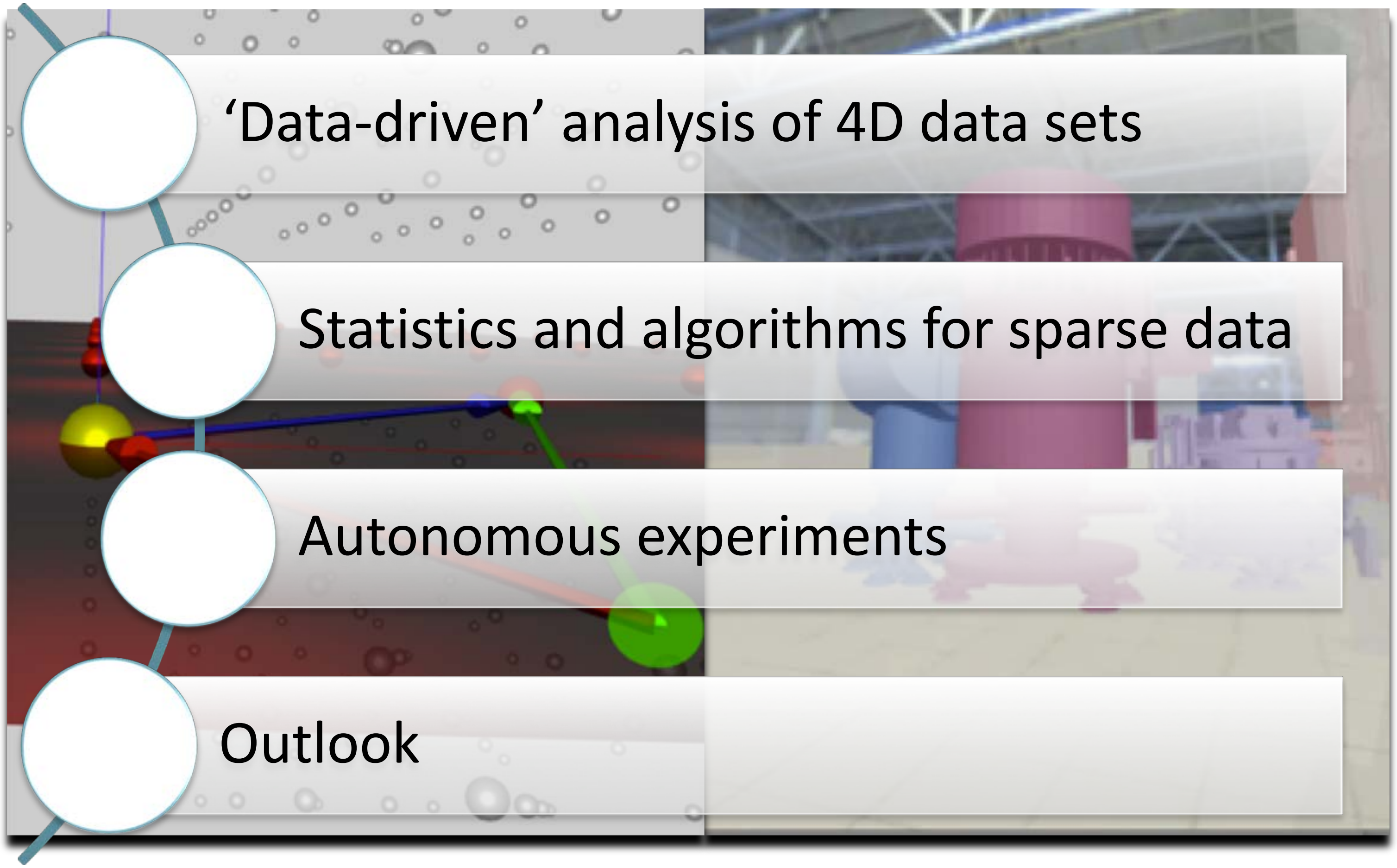


D. Jullien et al., Nucl.Instr & Meth A **1010** (2021), p. 165558



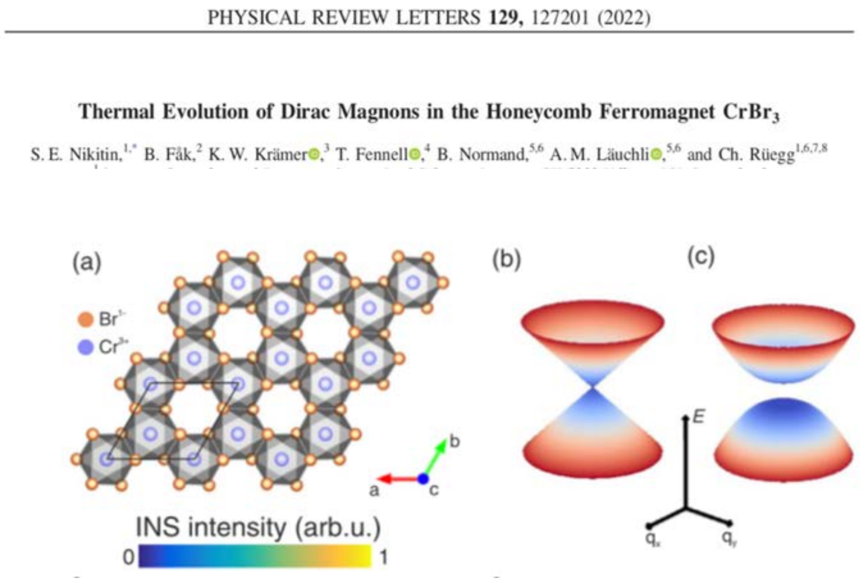
S.E. Nikitin et al., PRL **129** (2022), p. 127201

Autonomous Experiments in Inelastic Neutron Scattering



- ‘Data-driven’ analysis of 4D data sets
- Statistics and algorithms for sparse data
- Autonomous experiments
- Outlook

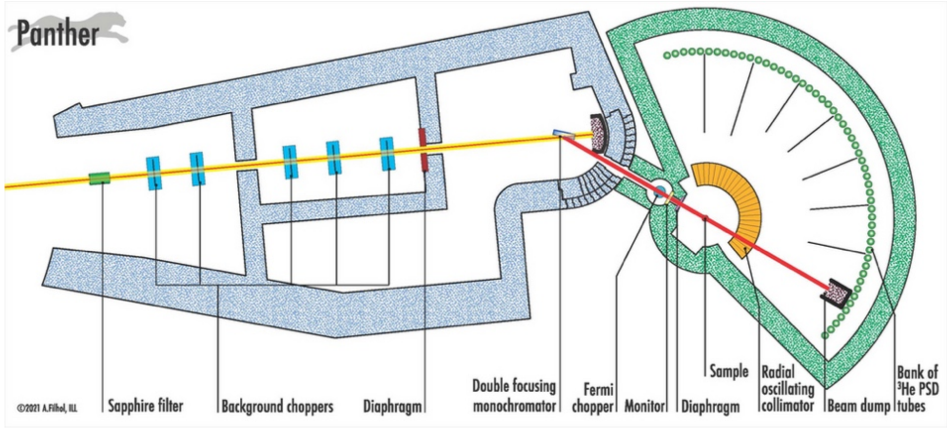
Exploring S(**Q**,ω) @ Panther:

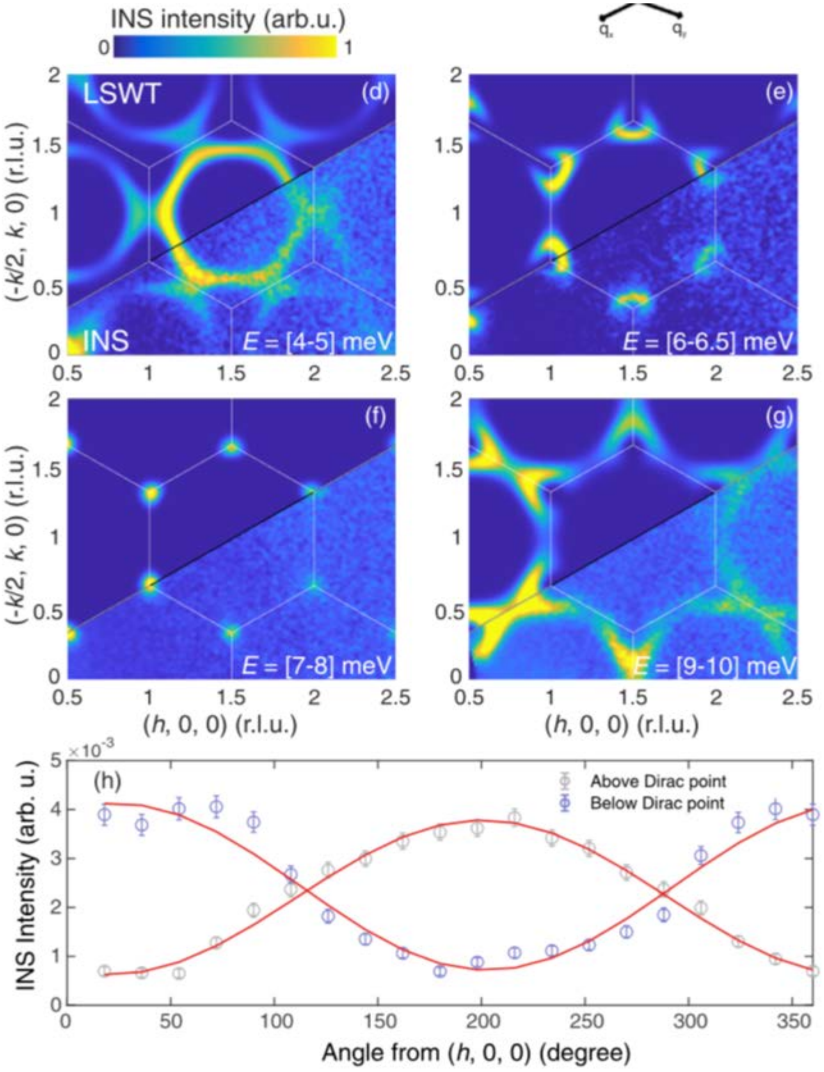


PHYSICAL REVIEW LETTERS **129,** 127201 (2022)

Thermal Evolution of Dirac Magnons in the Honeycomb Ferromagnet $CrBr_3$

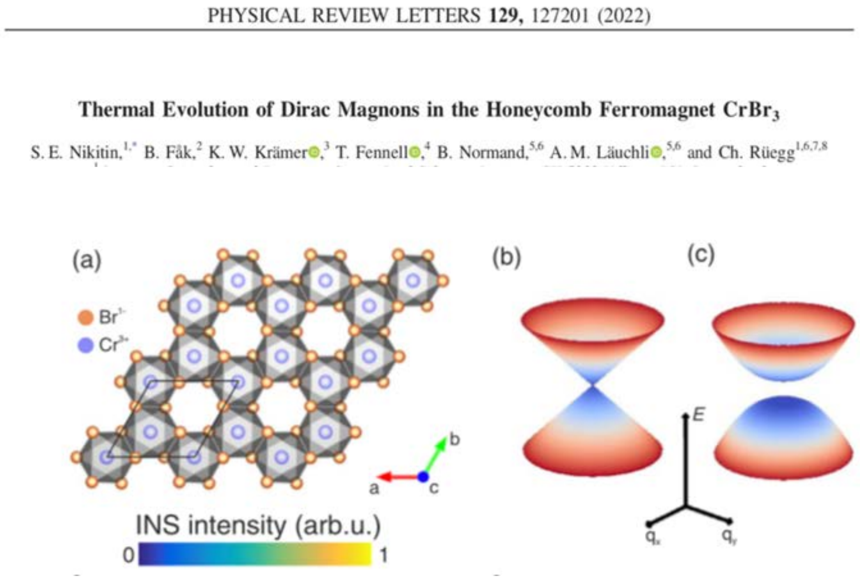
S. E. Nikitin,[1,*] B. Fåk,[2] K. W. Krämer,[3] T. Fennell,[4] B. Normand,[5,6] A. M. Läuchli,[5,6] and Ch. Rüegg[1,6,7,8]





Panther @ILL

Exploring S(Q,w) – fitting 4D data sets:



PHYSICAL REVIEW LETTERS **129,** 127201 (2022)

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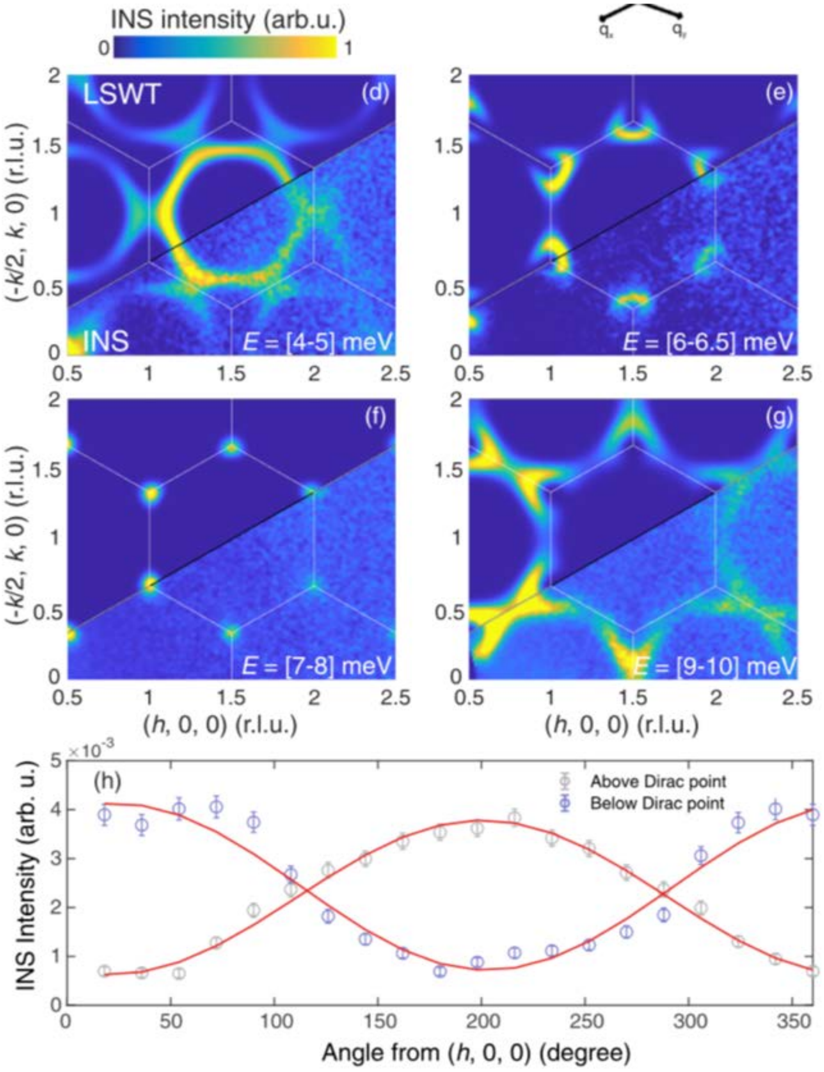


Panther @ILL

Simulation/ fitting:

- 139 constant-Q cuts
- Determination of J's
- Calculation of S(Q,w)

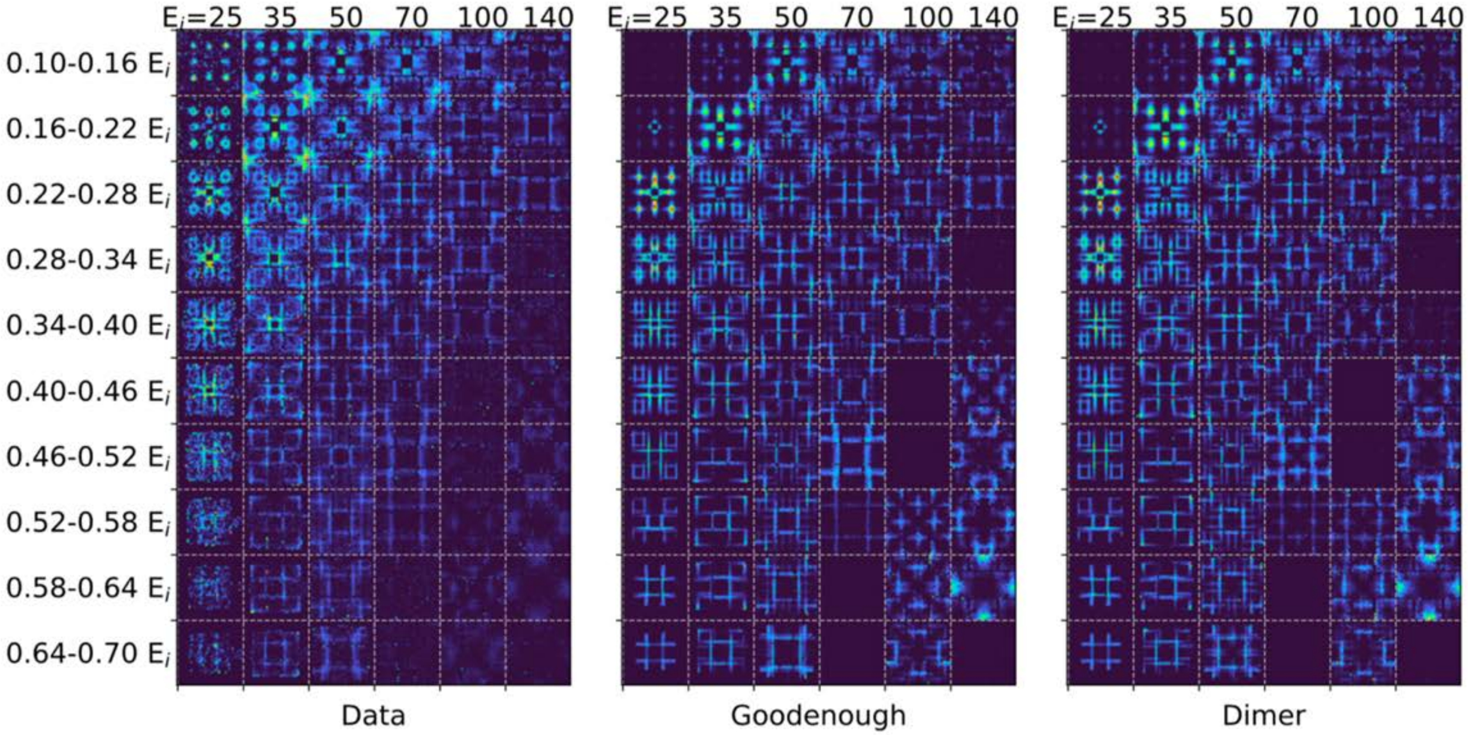
Horace **SpinW**

S. Toth and B Lake, *J. Phys.: Condens. Matter* **27** (2015) 166002.
R.A. Ewings et al., Nucl.Instr.Meth.A **834** (2016) 132.

Exploring S(**Q**,ω) via Machine Learning :

ML Neural network approaches – Differentiating models

MAPS @ ISIS Simulated data for training CNN



Data volume:
~ 30 million (**Q**,ω) values compressed to
400 x 240 pixel image

Simulation/Convolution:
6644 generated images (LSWT)
~ 7000 CPU-hr = 290 CPU-days
(~ several weeks on a cluster)

Comparison of several convolution methods

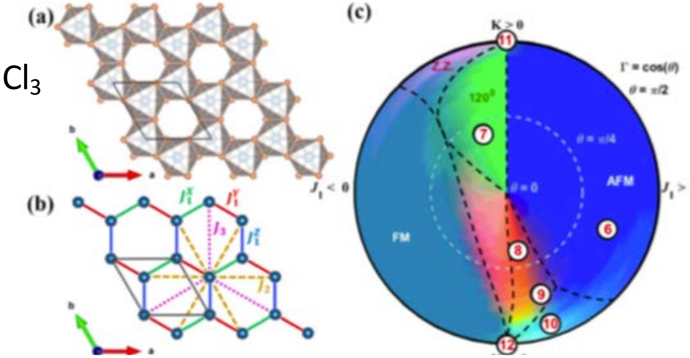
ML methods:
- CNNs
- Deterministic Uncertainty quantification (DUQ)
- Network complexity vs interpretability (Class activation maps CAMs).

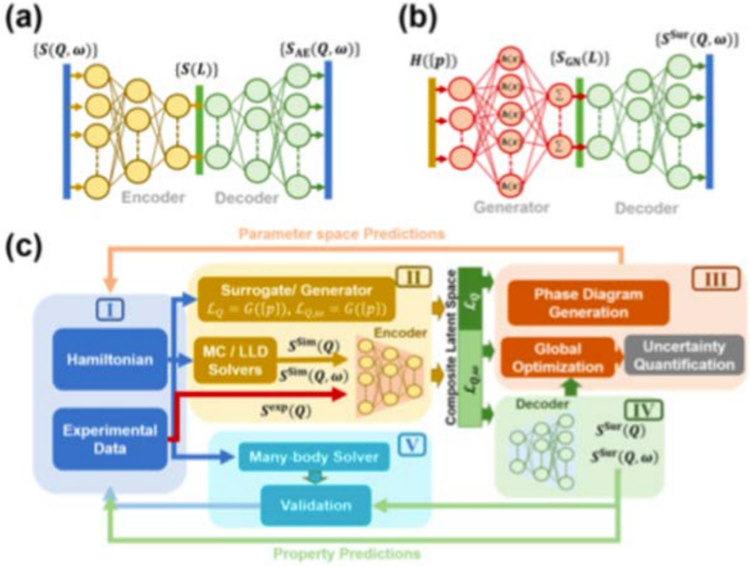
Keith T Butler *et al* 2021 *J. Phys.: Condens. Matter* **33** 194006.

Exploring S(Q,w) via Machine Learning:

Extraction of interaction parameters

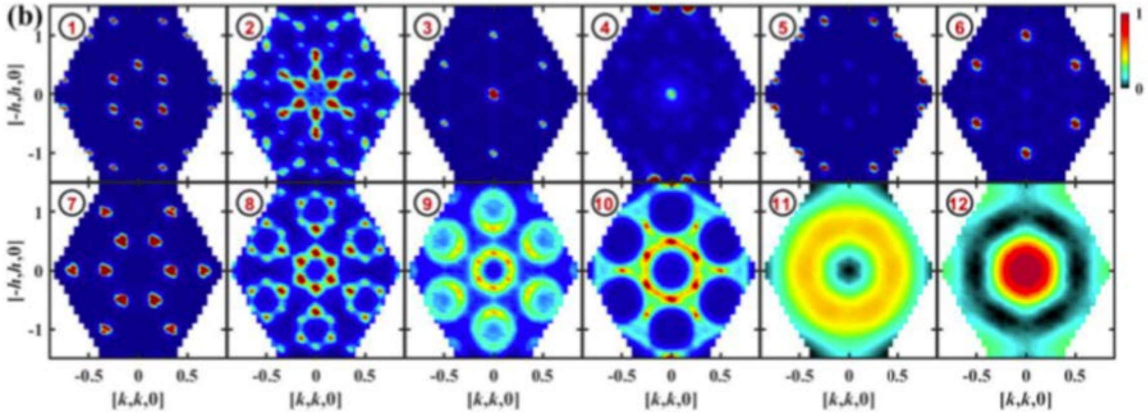
α-$RuCl_3$



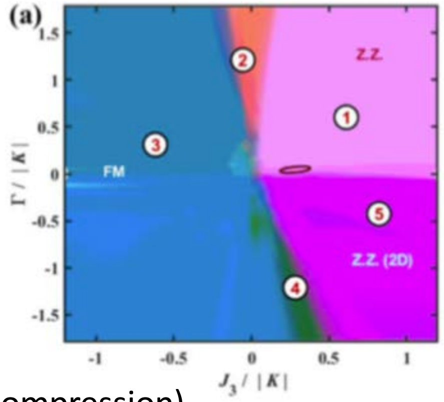


A.M. Samarakoon et al. , Phys.Rev.Res. **4** (2022), L022061

5 model parameters

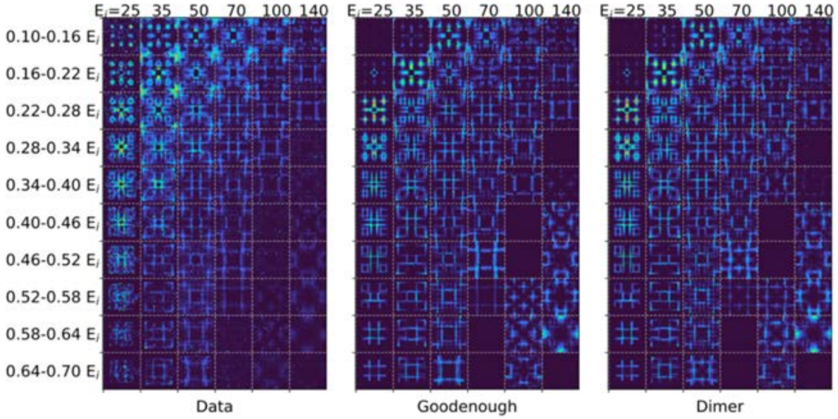


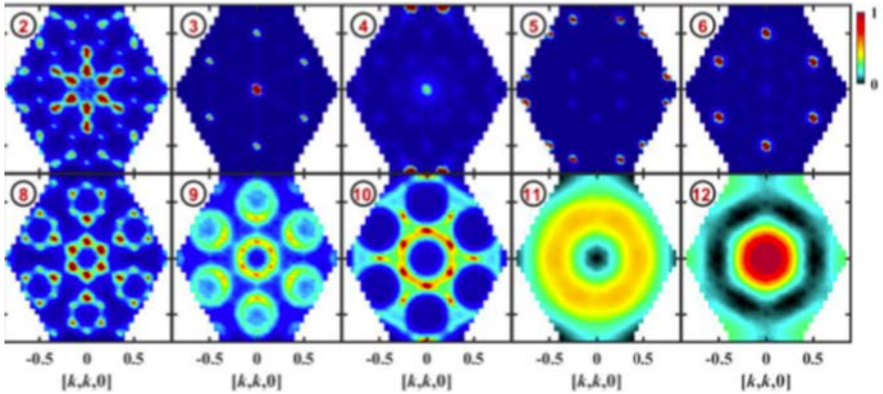
Simulation/Convolution:
10^4 models (MC/LLD solver)



ML methods:
- Nonlinear Autoencoder (denoising and compression)
- ML assisted iterative mapping algorithm (IMA)
- Radial basis networks for fast surrogates

Exploring S(**Q**,ω) via 'data driven' methods:





Specifically trained NN for well posed questions

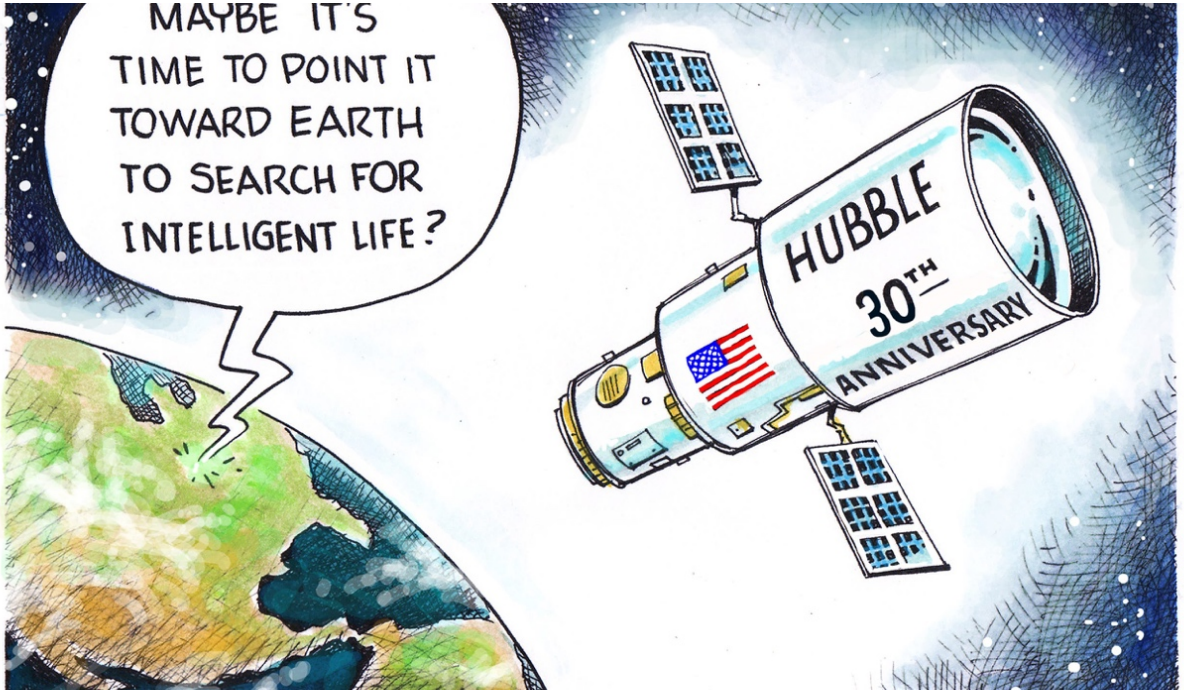
A few, but huge data sets (~ Tbytes)

Huge computational/simulation effort

Benefits:

Acceleration of data treatment
Denoising

Exploring S(**Q**,ω) – from the TAS perspective:



limited solid angle

limited experimental budget = NoP

flexible exploration

asking the right questions.

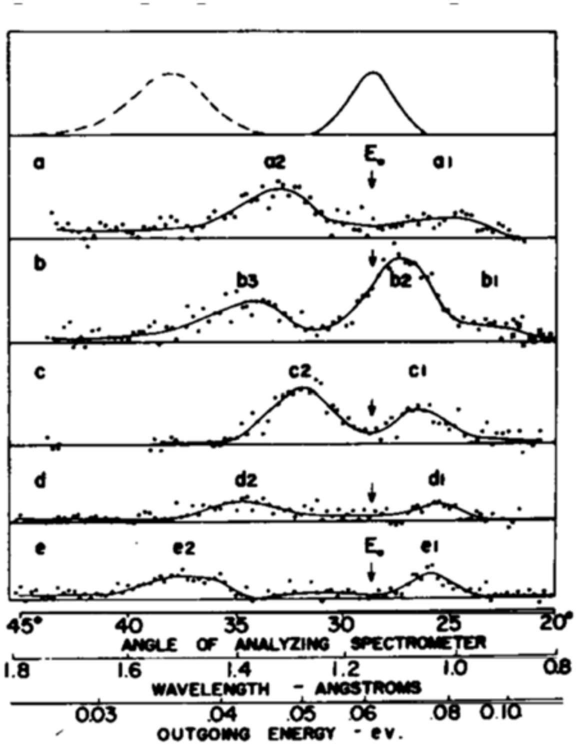
asking the right questions *before* the experiment.

How many data do we need?
Can we use our experimental budget better?

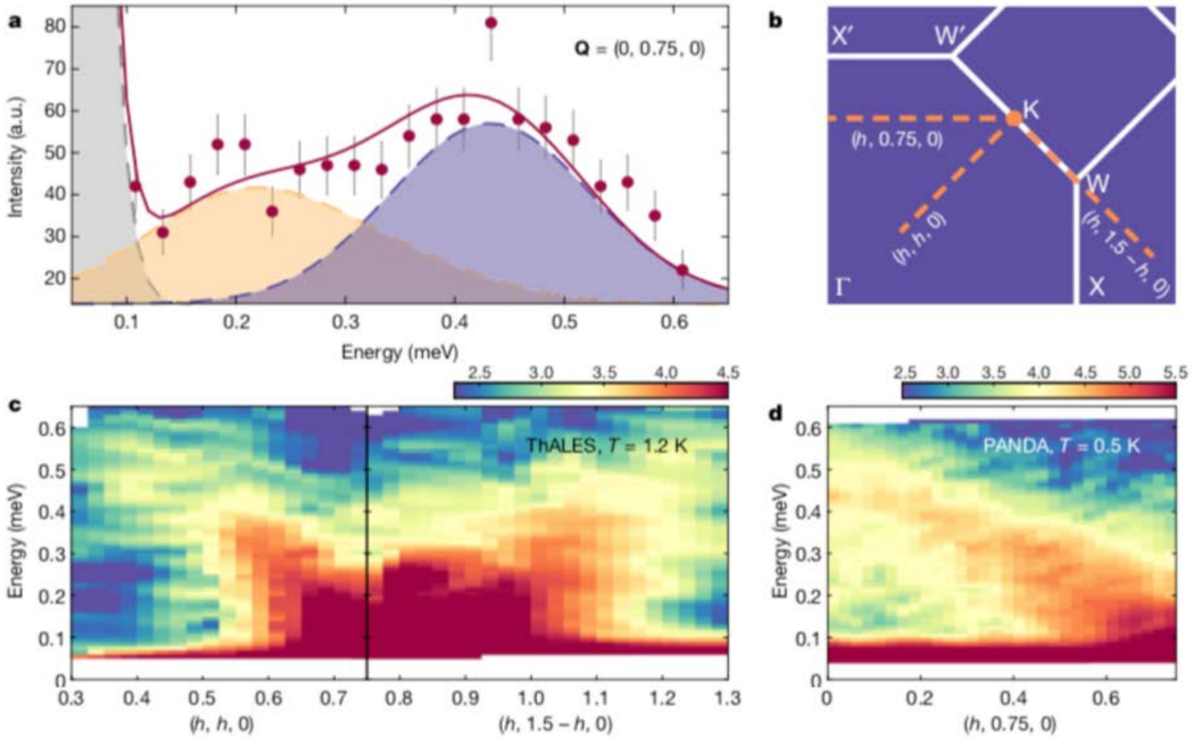
A different view on:
-) Statistics
-) Acquisition strategy

TAS: From 1954 to today

Establishment of const - (**Q** or E)- scans



Brockhouse B.N. and Stewart A.T. *Scattering of Neutrons by Phonons in an Aluminum Single Crystal*. Phys. Rev. 100:756 - 757, **1955**



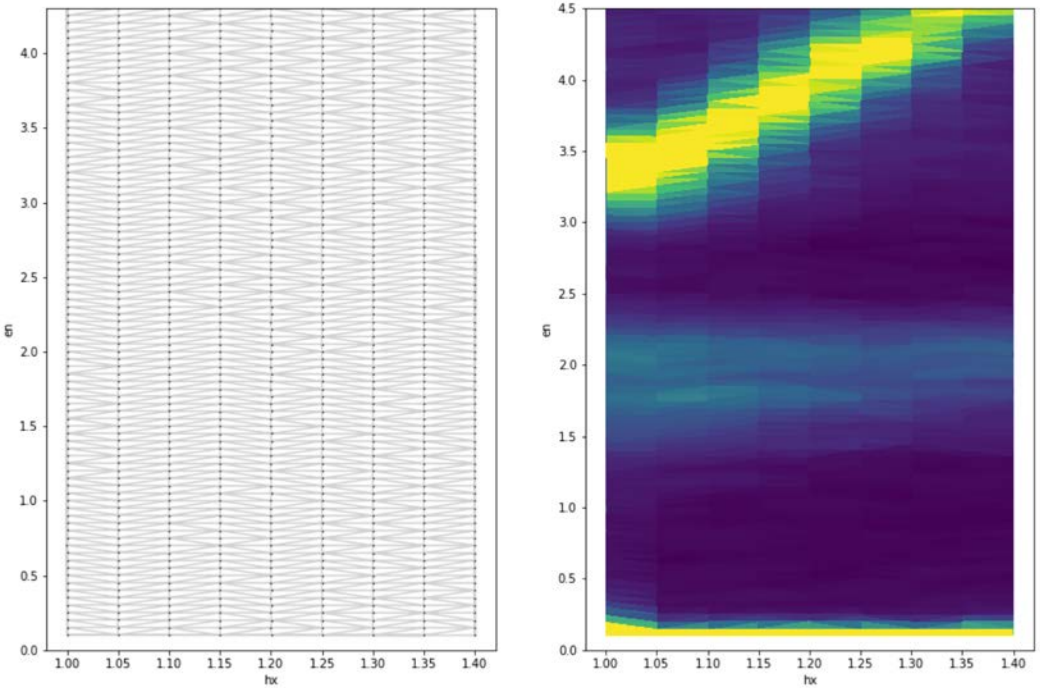
Gao, S. et al., *Nature* **586,** 37–41 (2020)

Exploring S(Q,w) – correlating points:

Can we use our experimental budget better?

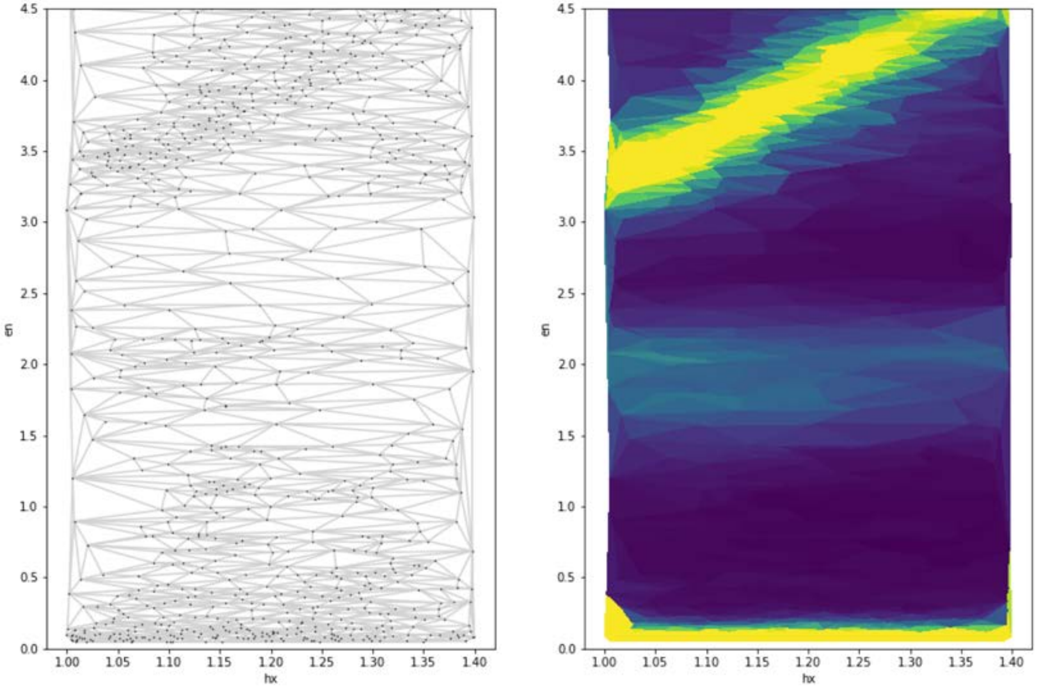
Grid scanning:

constant energy scans

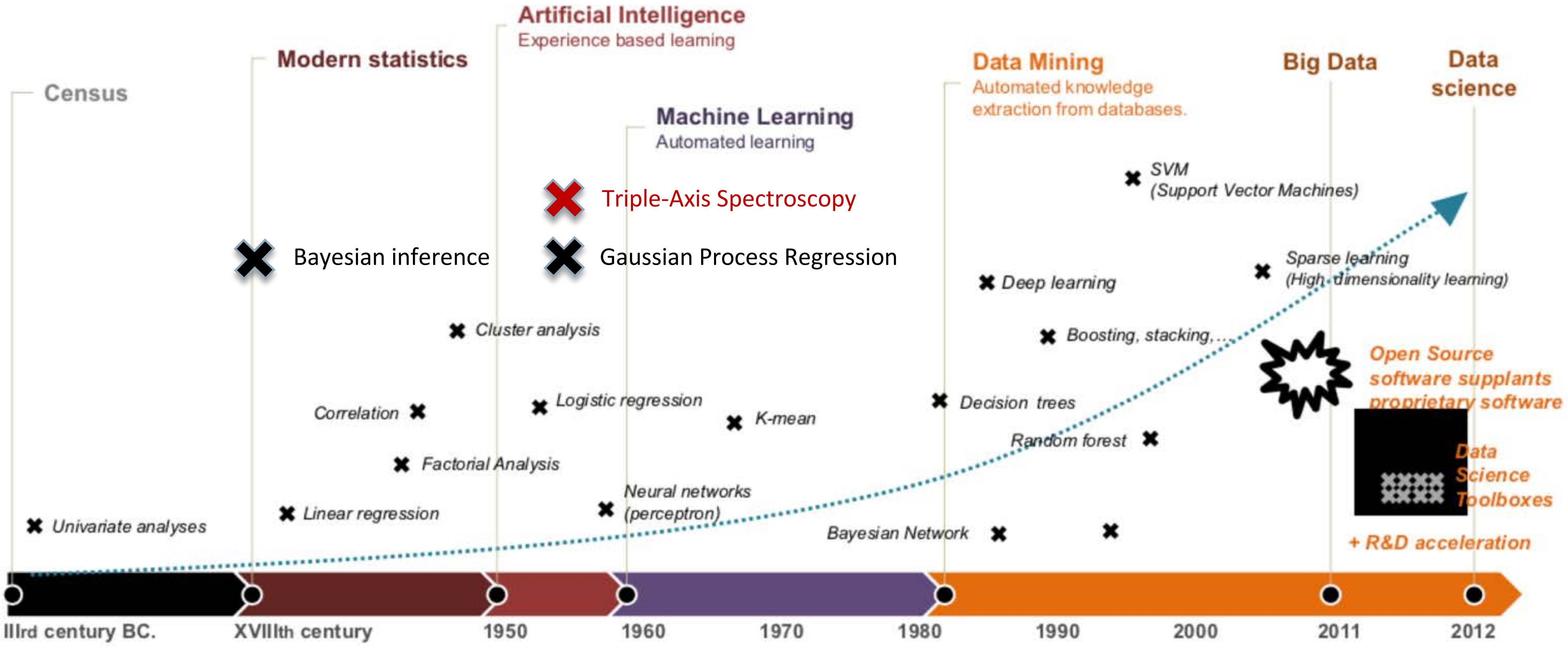


Algorithm driven scanning:

Gaussian Process Regression



‘New’ algorithms – a look back into statistics:

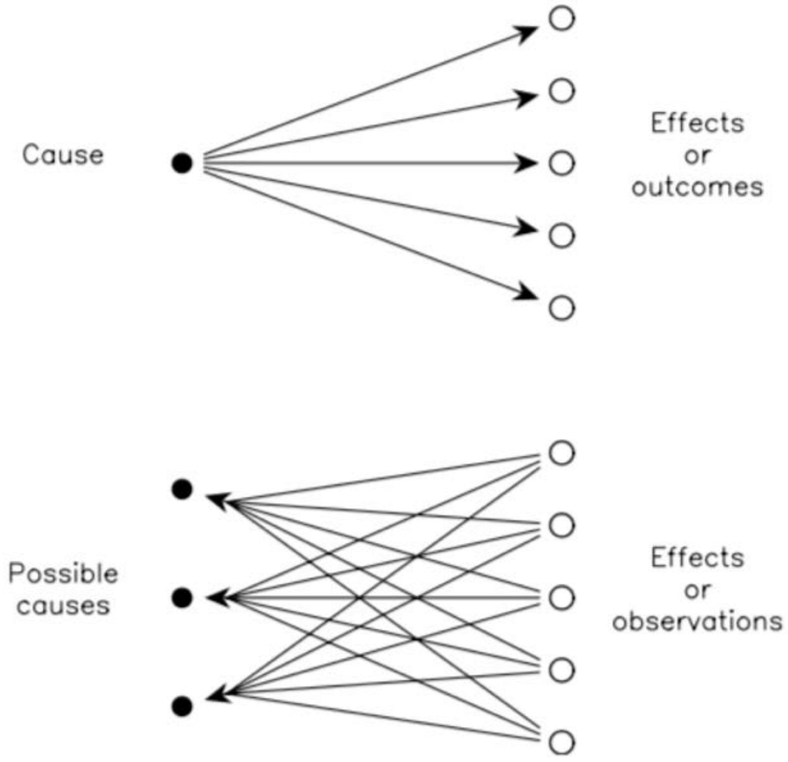


A. Nesvijevskaia. Phénomène Big Data en entreprise, Science de l’information et de la communication. Conservatoire national des arts et metiers – CNAM, 2019. NNT: 2019CNAM1247.

Bayesian inference – a probabilistic method:

"We owe to the frailty of the human mind one of the most delicate and ingenious of mathematical theories, namely the science of chance or probabilities." Pierre Simon Laplace[*)]

Thomas Bayes (1701 – 1761)
Pierre Simon Laplace (1749 – 1827)



$$P(\Theta|y) = \frac{P(y|\Theta)P(\Theta)}{P(y)}$$

[*)] *The theory that would not die*, Sharon B. McGrayne, Yale University Press, 2011.

Bayesian inference – a probabilistic method:

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Thomas Bayes (1701 – 1761)
Pierre Simon Laplace (1749 – 1827)

Gravitational waves:

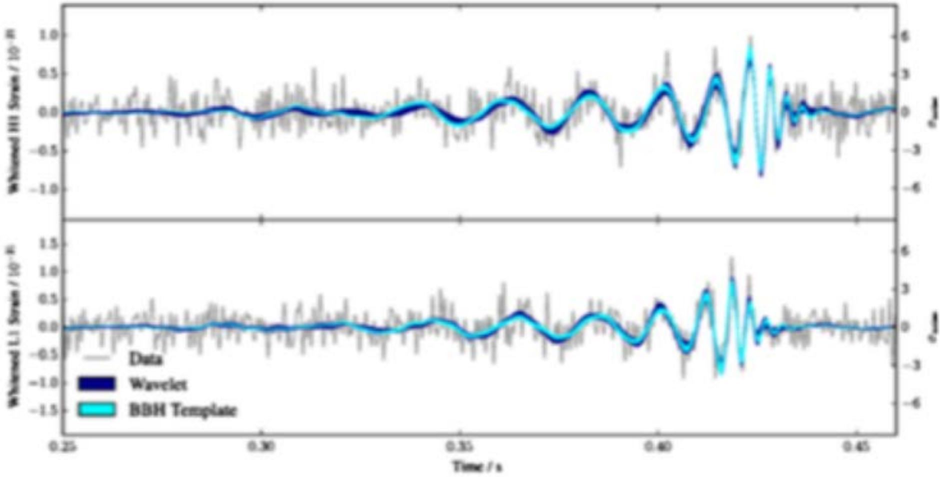


FIG. 1. Measured detector strain time series of the first detected gravitational wave signal by Advanced LIGO, GW150914 (B. P. Abbott *et al.*, 2016b) as observed in H1 (top panel) and L1 (bottom panel). The times displayed are with respect to September 14, 2015, 09:50:45 U

is are

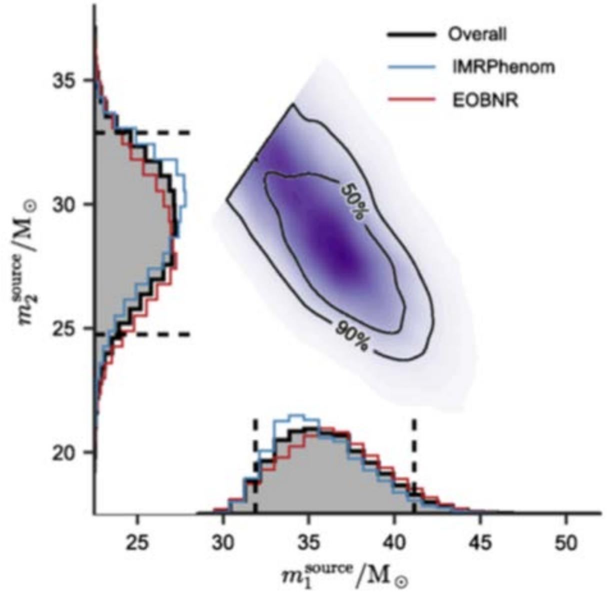


FIG. 2. Source frame component mass parameter posterior probability distributions for the first detected gravitational wave signal by Advanced LIGO, GW150914 (B. P. Abbott *et al.*,

N. Christensen and R.Meyer, *Parameter Estimation with gravitational waves*, Rev.Mod.Phys. 94 (2022), 025001

Gaussian Process Regression:

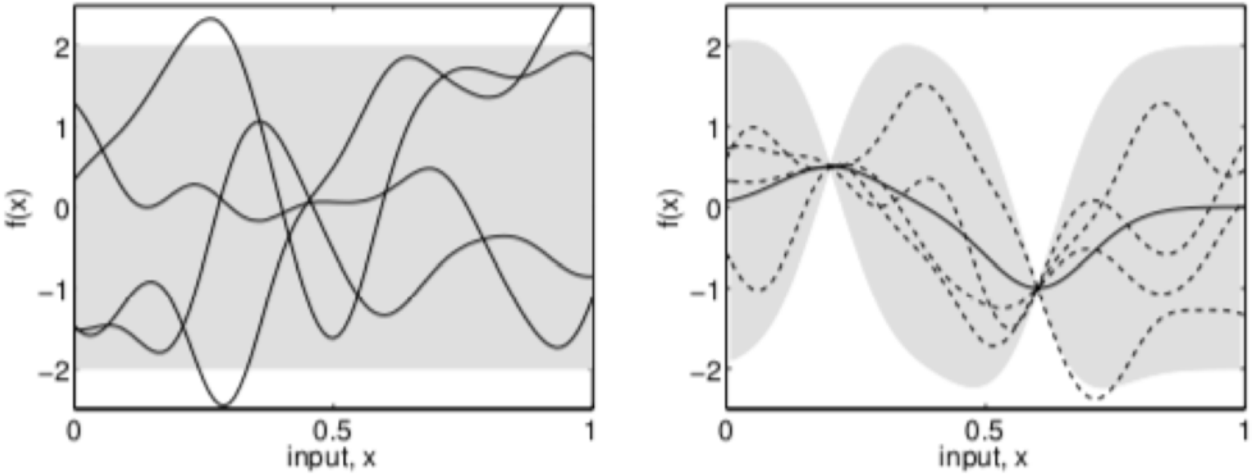
Definition: A Gaussian process is a collection of random variables, any finite number of which have a joint Gaussian distribution. 1)

$$y_j = f(\mathbf{x}_j) + \varepsilon_j, \qquad \varepsilon_j \sim \mathcal{N}(0, \sigma_n^2)$$

Prior distribution:

$$p(\mathbf{f} | \{\mathbf{x}_i\}_{i=1}^n) = \mathcal{N}(\mathbf{f} | \mathbf{0}, \mathbf{K_{ff}})$$

$$[\mathbf{K_{ff}}]_{ij} = k(\mathbf{x}_i, \mathbf{x}_j) = \mathbb{E}[f(\mathbf{x}_i) f(\mathbf{x}_j)]$$



(a), prior (b), posterior

Predictive distribution:

$$p(y_* | \mathbf{x}_*, \mathcal{D}) = \mathcal{N}(\mu_*, \sigma_*^2), \text{ where } \begin{cases} \mu_* = \mathbf{k}_{*\mathbf{f}}(\mathbf{K_{ff}} + \sigma_n^2 \mathbf{I}_n)^{-1}\mathbf{y} \\ \sigma_*^2 = \sigma_n^2 + k_{**} - \mathbf{k}_{*\mathbf{f}}(\mathbf{K_{ff}} + \sigma_n^2 \mathbf{I}_n)^{-1}\mathbf{k}_{\mathbf{f}*} \end{cases}$$

1)C.E. Rasmussen & K.I. Williams, *Gaussian Processes for Machine Learning*, MIT Press, 2006.

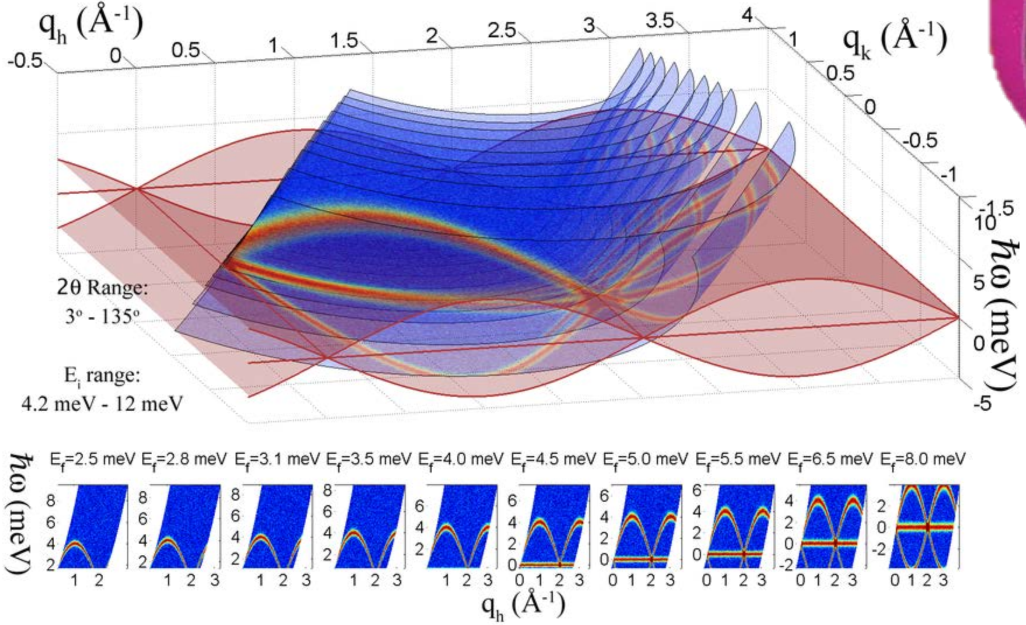
M. Lázaro-Gredilla et al., Sparse Spectrum Gaussian Process Regression, J. Mach. Learn. Res. 11 (2010), 1865-1881.

Kriging Regression :

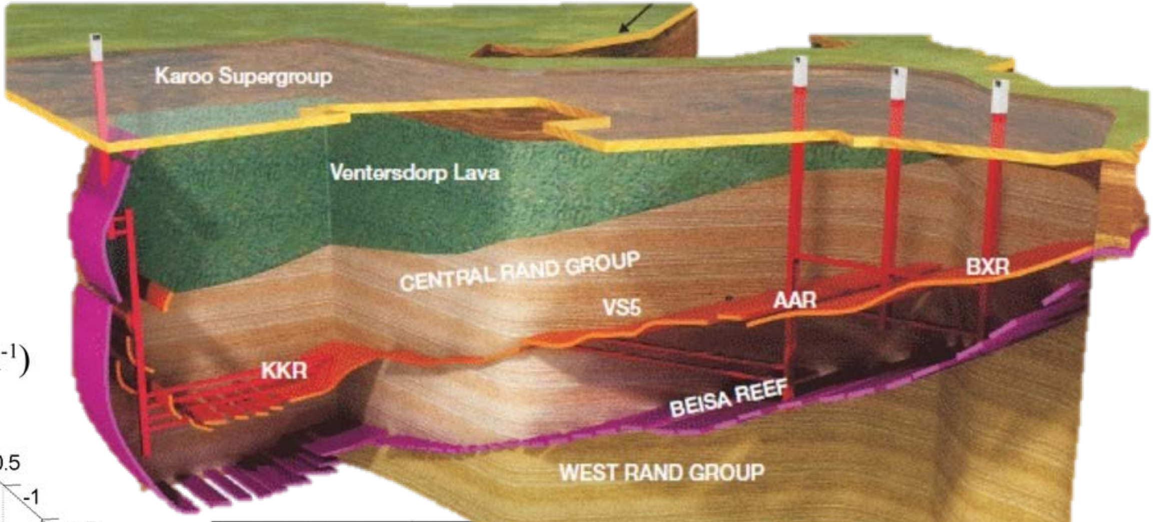
Daniel G. Krige: distance-weighted average gold grades at the Witwatersrand reef (South Africa).

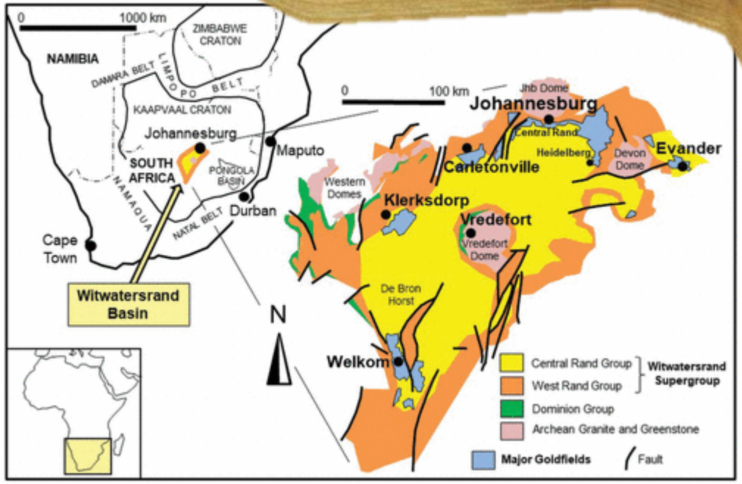
Georges Matheron: mathematical framework 1960.

1g Au = 8 min TAS

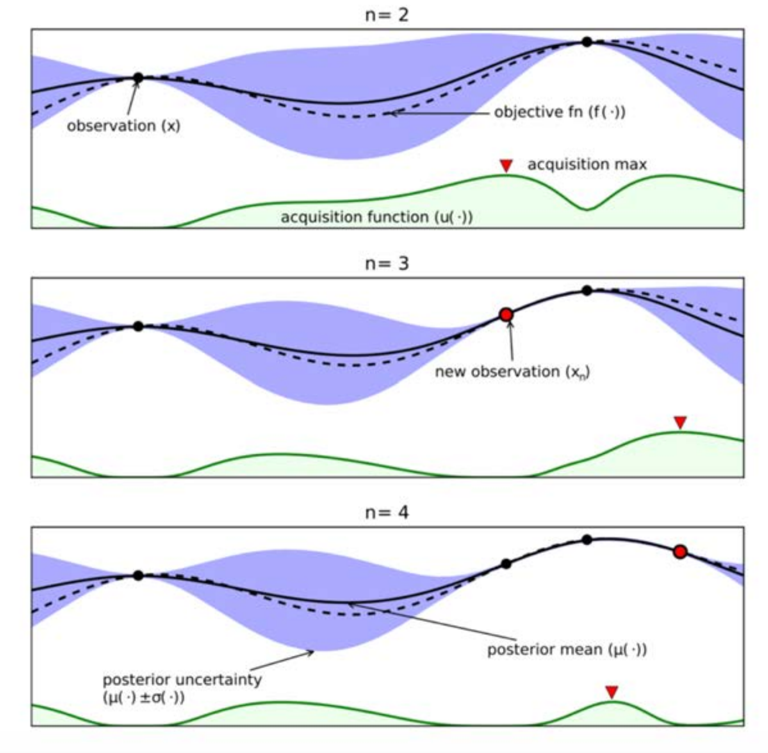


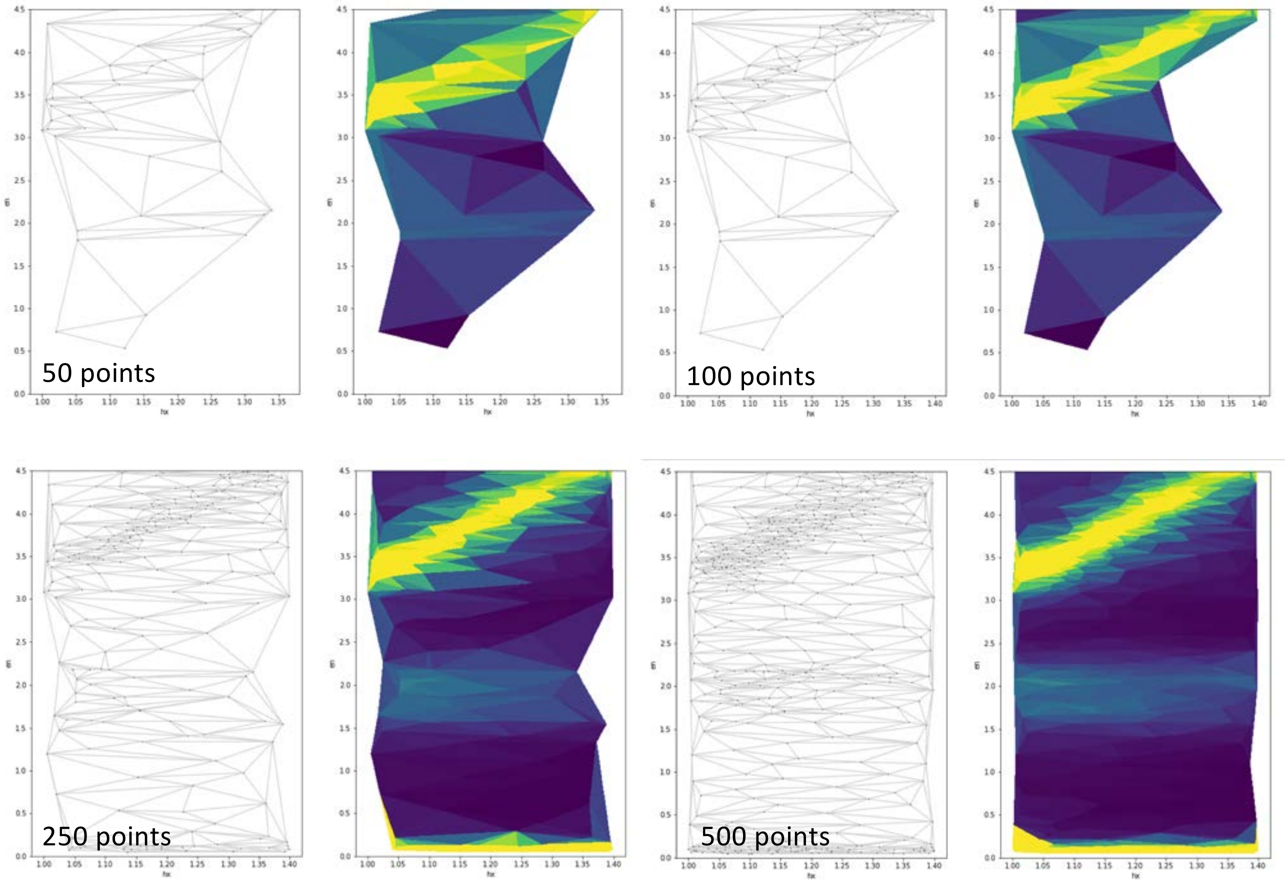
© J.O. Birk and the BIFROST team @ESS, Lund





Bayesian optimization – Steering the position:





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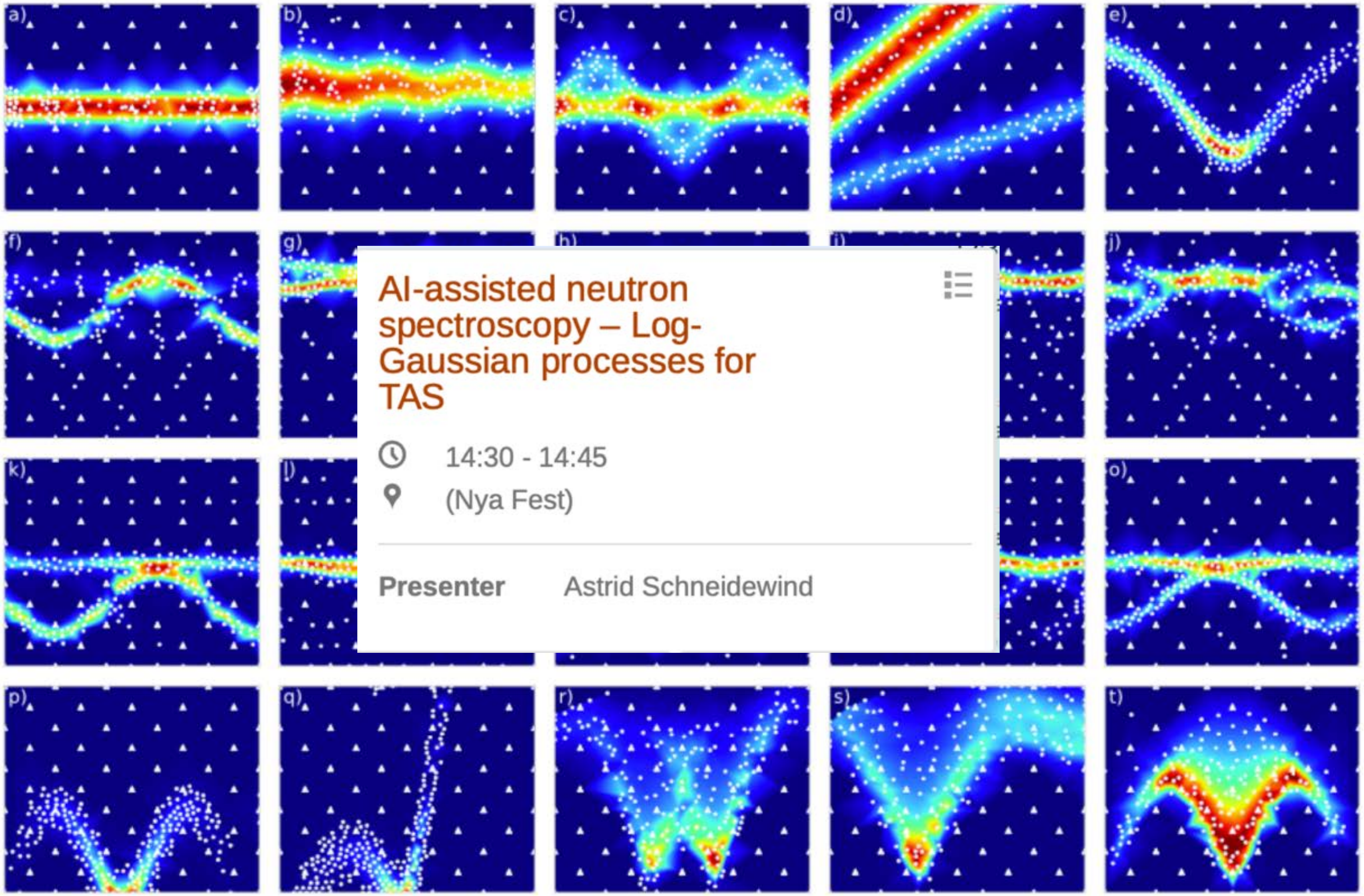
[1]) B. Shahriari et al., *Taking the Human out of the Loop: A review on Bayesian optimization*, Proceedings of the IEEE 104 (2016), p.148

Log - GPR:



AI-assisted neutron spectroscopy – Log-Gaussian processes for TAS

14:30 - 14:45

(Nya Fest)

Presenter Astrid Schneidewind

M. Teixeira Parente et al., *Log-Gaussian processes for AI-assisted TAS experiments*, arXiv:2209.00980v1

Towards Autonomous Experiments:

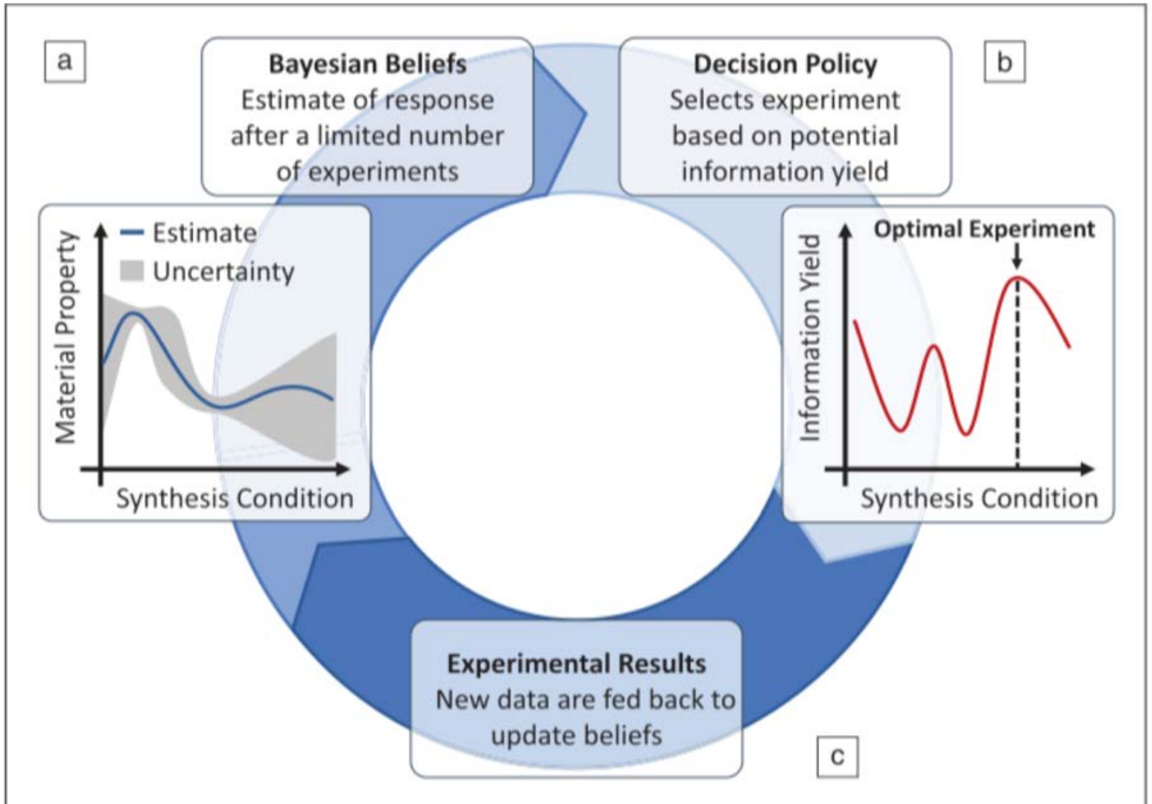
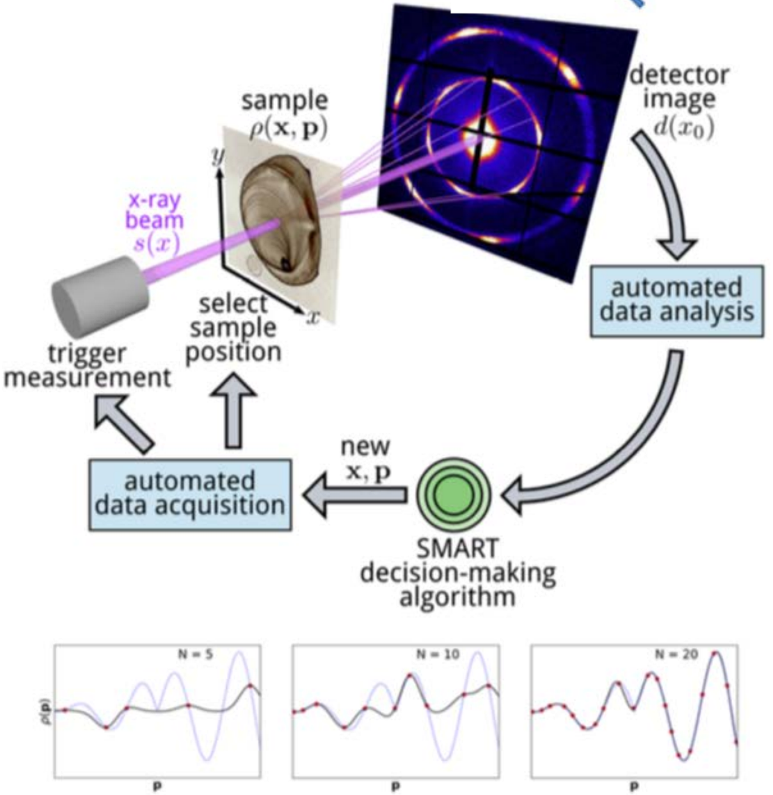


Figure 2. The closed-loop planning, execution, and learning from experiments for guided and autonomous research. (a) Beliefs about the system that are captured using Bayesian statistics, estimating a material property of interest; (b) a decision policy that balances exploration versus exploitation when selecting an experiment to run; (c) closing the loop by using the Bayesian update.

K.G. Reyes and B. Maruyama, MRS Bull. 44, 530 (2019)

https://gpcam.lbl.gov



Noack, M.M., *et al.* *Sci Rep* **9,** 11809 (2019)

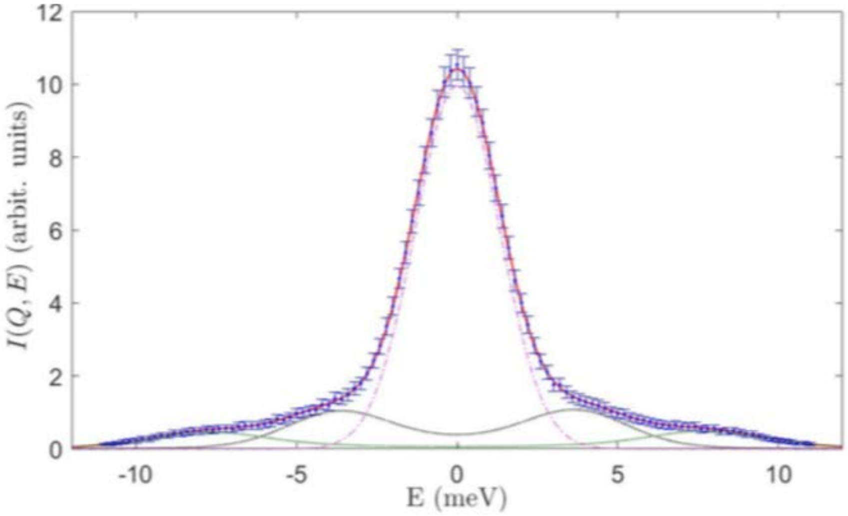
Intermediate conclusions:

- Single crystal spectroscopy!
- Complex experiments impose complex data analysis
- ML algorithms arrived also in the INS community (data driven and data optimization)
- Intelligent robotic reciprocal space explorer
- *Bayesian optimization is the future*

Physics instructed AE with Brillouin data:

Bayesian optimization on Brillouin scattering data

$$S(Q,E) = A_e(Q)\delta(E) + [n(E)+1]\frac{E}{k_BT}\left\{\sum_{k=1}^{2}\frac{2}{\pi}A_k(Q)DHO_k(Q,E)\right\}$$



A. DeFrancesco et al., PRE 94, 023305 (2016)

Quasi-elastic neutron scattering

D.S. Sivia et al., Physica B **182** (1992), 341
D.S. Sivia et al., *Data Analysis – a Bayesian Tutorial*, Oxford University Press 2006.

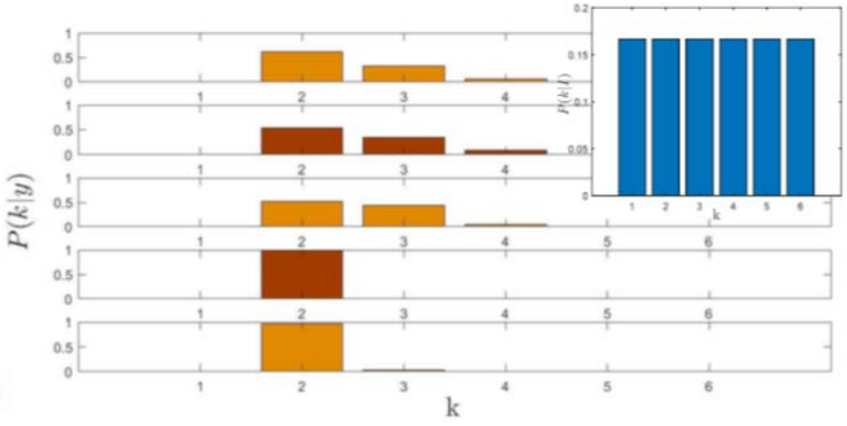
NSE

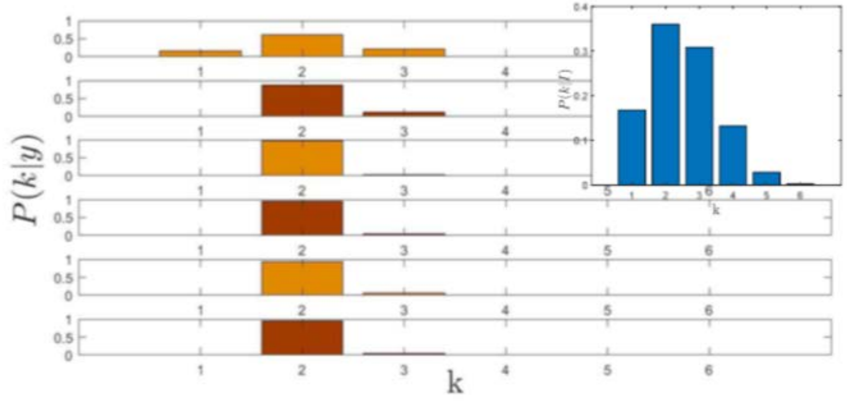
A. De Francesco et al., PRE **99**, 052504 (2019)

Autonomous loop

A. De Francesco et al., IntechOpen. https://doi.org/10.5772/intechopen.103850

AE with Brillouin data - parameter estimation :





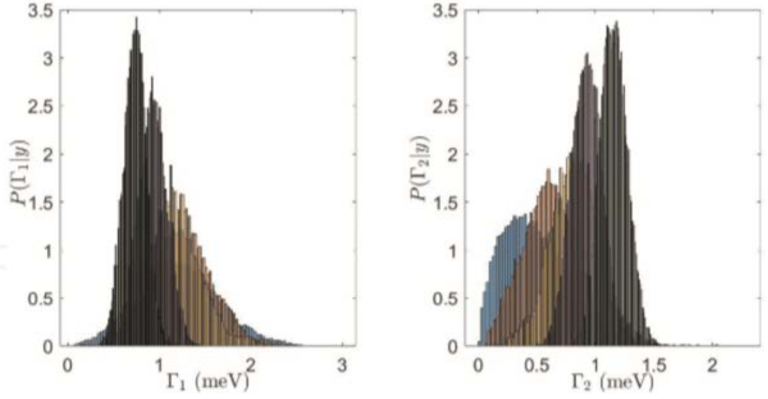


Figure 5.
Posterior distribution of the dampings of the two excitations as estimated from the Bayesian analysis after 5, 10, 20, 40, and 60 experimental runs.

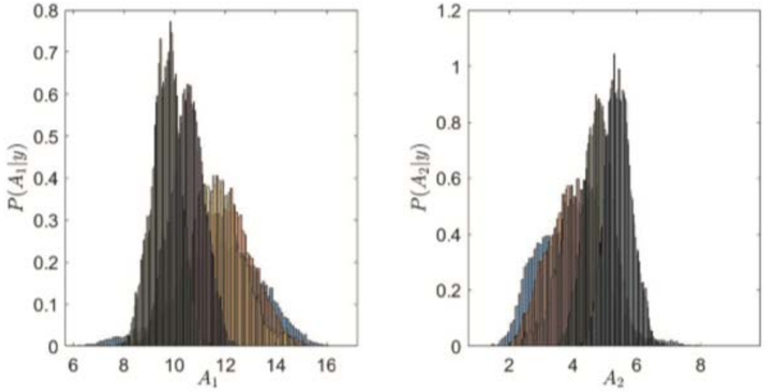


Figure 6.
Posterior distribution of the amplitudes of the two excitations estimated from the Bayesian analysis after 5, 10, 20, 40, and 60 experimental runs.

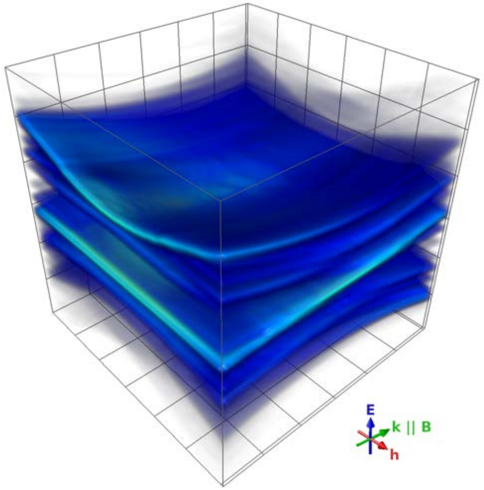
A. De Francesco, L. Scaccia, M. Boehm and A. Cunsolo, *Bayesian inference as a tool to optimize spectral acquisition in scattering experiments*, IntechOpen. https://doi.org/10.5772/intechopen.103850

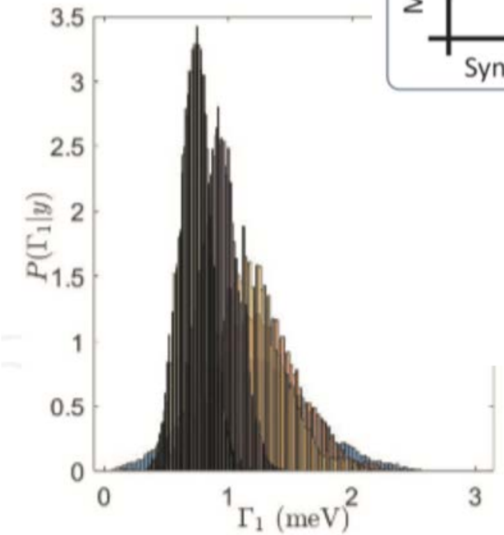
Towards Physics Informed Autonomous Experiments:

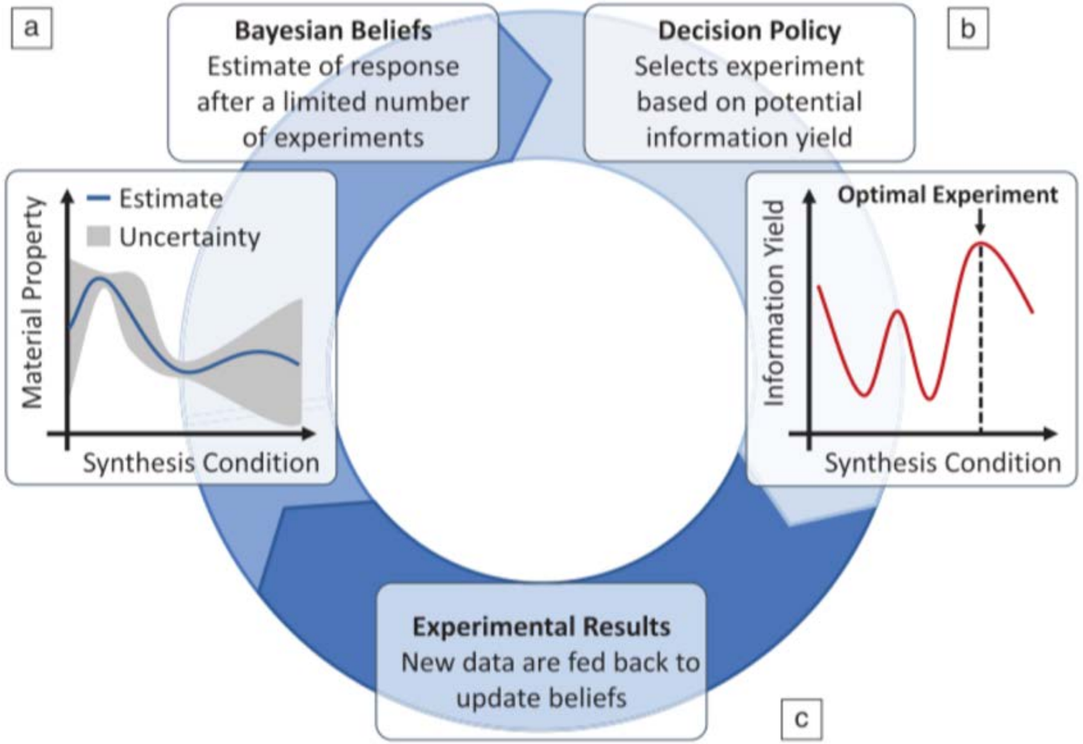
$H_1(\boldsymbol{Q},\omega,J_1,..,J_N)$

$H_2(\boldsymbol{Q},\omega,J_1,..,J_M)$

$(\mathbf{Q_0},\omega_0)$





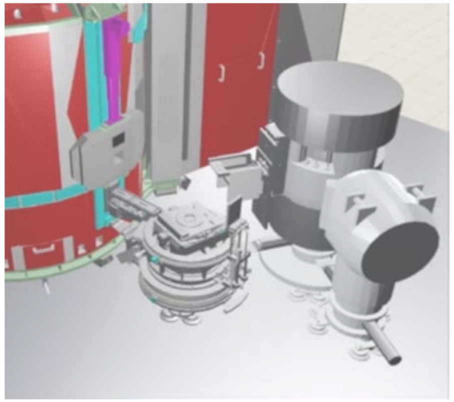


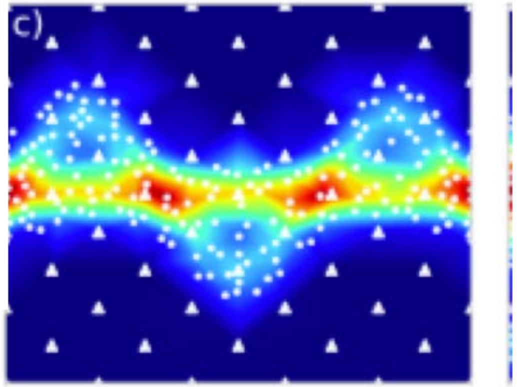
Bayesian Beliefs
Estimate of response after a limited number of experiments

Decision Policy
Selects experiment based on potential information yield

Experimental Results
New data are fed back to update beliefs

K.G. Reyes and B. Maruyama, MRS Bull. 44, 530 (2019)





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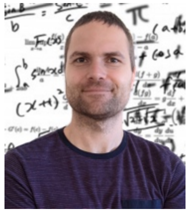

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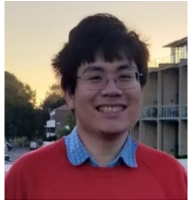

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Thank you!