High resolution inelastic neutron scattering

PELICAN & EMU

Dr Gail N. Iles
Lecturer in Physics
School of Science

RMIT UNIVERSITY

Acknowledgement of Country

I acknowledge the Dharawal speaking people, traditional custodians of the land, and pay my respects to elders past and present.

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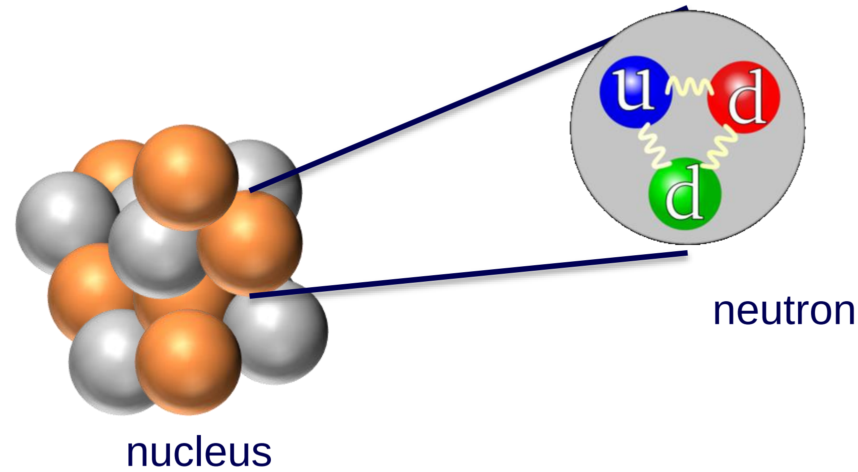
Overview

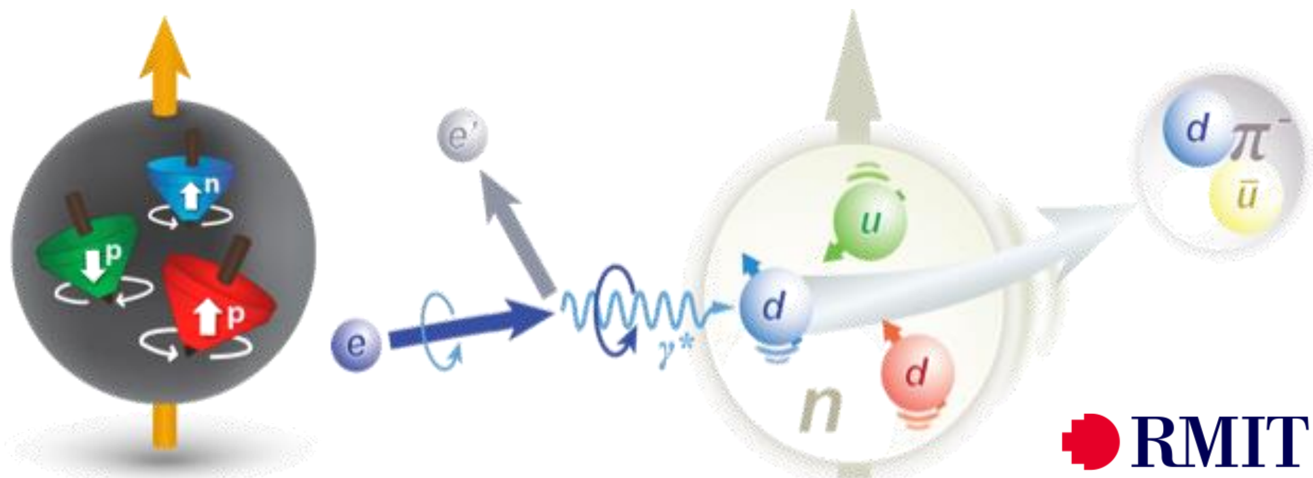
1. Neutron scattering
   a. Neutron properties
   b. Terminology
   c. Cross sections and the scattering function
2. Inelastic neutron scattering
   a. Inelastic data
   b. kinematics
3. Time-of-flight neutron spectroscopy
4. Backscattering neutron spectroscopy

1. Neutron Scattering

Neutrons

- Neutrons are subatomic particles
  - symbol: n or n^0
  - mass = 1.6749×10^{-27} kg
  - no net electric charge
  - high penetration depth
  - have a magnetic moment
    - quark substructure
    - spin = ½
    - fermions





Physics at RMIT

- de Broglie **wavelength** of the neutron $$\lambda = \frac{h}{mv}$$ (direction being that of its velocity)

- **Kinetic energy** of slow neutrons with velocity **v** $$E = \frac{1}{2}mv^2$$

- **Wavevector *k*** of the neutron has magnitude $$k = \frac{2\pi}{\lambda}$$

- **Momentum** of neutron $$p = \hbar k$$ where $$\hbar = \frac{h}{2\pi}$$

Units

- In neutron scattering you will often find the **same** properties reported with ***different*** units
- Get used to converting between units (not all techniques use SI)
- Become familiar with **approximate** conversion rates
  - Energy, E (J, eV)
  - Wavelength, λ (nm, Å)
  - Optical frequency, f (Hz)
  - Angular frequency, ω (Hz)
  - Velocity, v (ms^{-1})
  - Wave vector, k ($Å^{-1}$, cm^{-1})
  - Temperature, T (K, °C, F)

A neutron at room temperature possesses 25 meV of energy

Using: $E = k_B T$

Thermal neutrons	@ 293 K	= 25 meV
Cold neutrons	@ 20 K	= 2 meV

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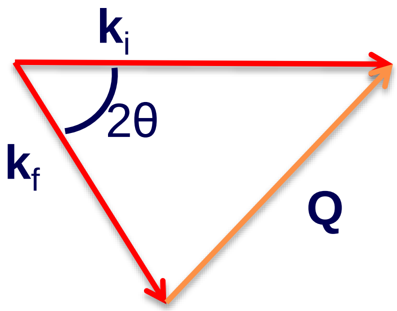
Wave vector, k

- A neutron with incident wave vector $\mathbf{k}_i$, interacts with a sample

- The neutron's outgoing wave vector is $\mathbf{k}_f$
- $\mathbf{k}_f$ makes an angle 2θ to $\mathbf{k}_i$

Scattering vector

- In reciprocal space, we create the scattering triangle



RECIPROCAL Space

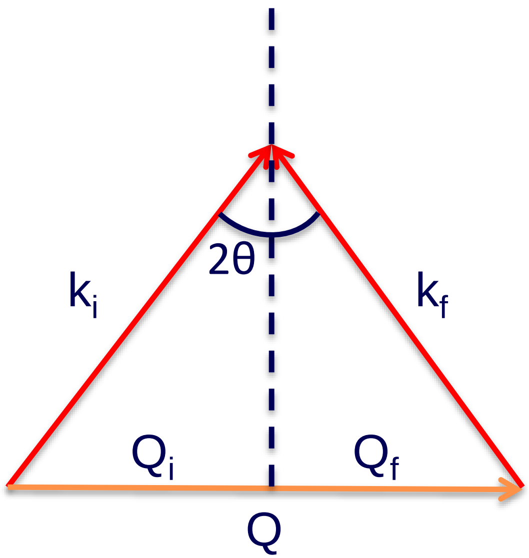
- Scattering vector, $\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f$
- Q denotes the momentum transfer

Reciprocal space is the **Fourier transform** of real space

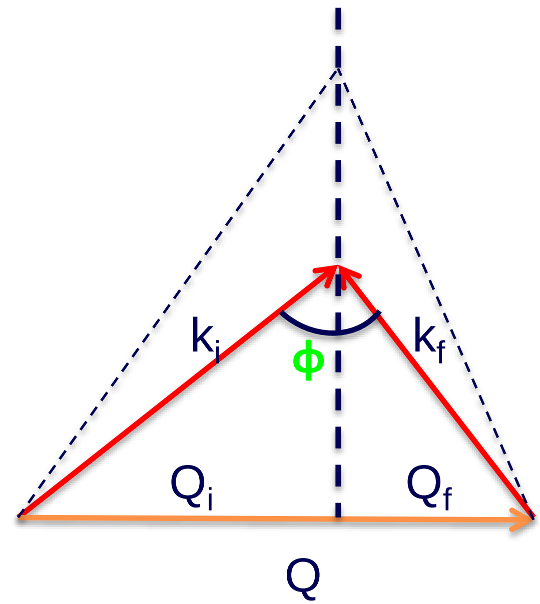
Momentum Transfer, Q

- Reciprocal space scattering diagram

Elastic case



Inelastic case



Elastic case: $Q_i = Q_f$

Inelastic case: $Q_i \neq Q_f$

Here, $Q_i > Q_f$ so momentum was given to the system

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Energy Transfer – ħω

- In terms of energy: $E_i = \frac{\hbar^2}{2m_n} k_i^2$ $E_f = \frac{\hbar^2}{2m_n} k_f^2$ where $\hbar = \frac{h}{2\pi}$

- Energy transfer: $\hbar\omega = E_i - E_f = \frac{\hbar^2}{2m_n}\left(k_i^2 - k_f^2\right)$ where $\omega = 2\pi f$

- Combining equations for energy and momentum transfer:

$$Q^2 = k_i^2 + k_f^2 - 2k_i k_f \cos\varphi$$

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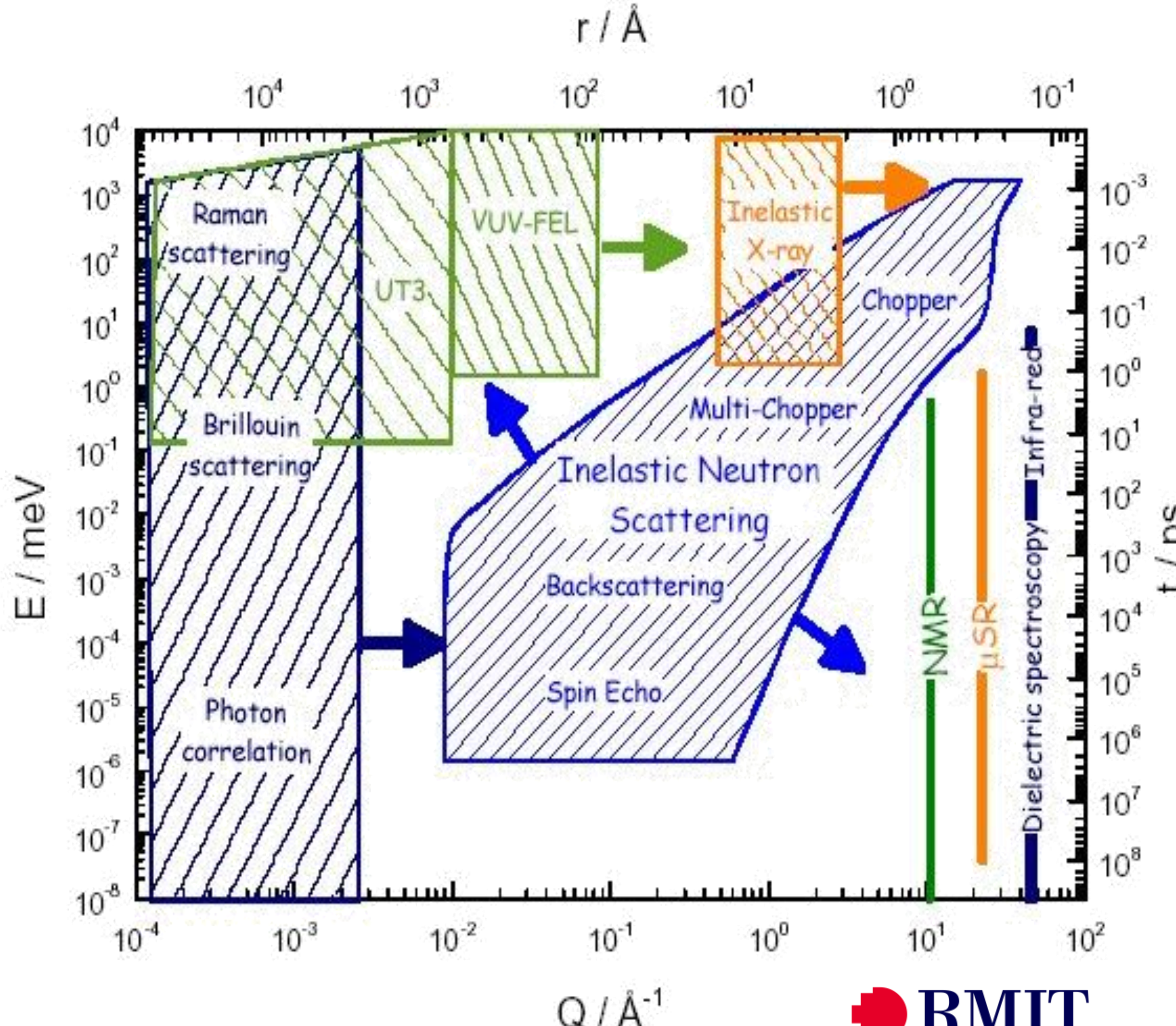
Probes of Condensed Matter

- Dynamical ranges
  - Real space
    - (r , t)
  - Reciprocal space
    - (Q,ω)
- Neutron scattering
  - Cross section
  - Energy
  - Temperature



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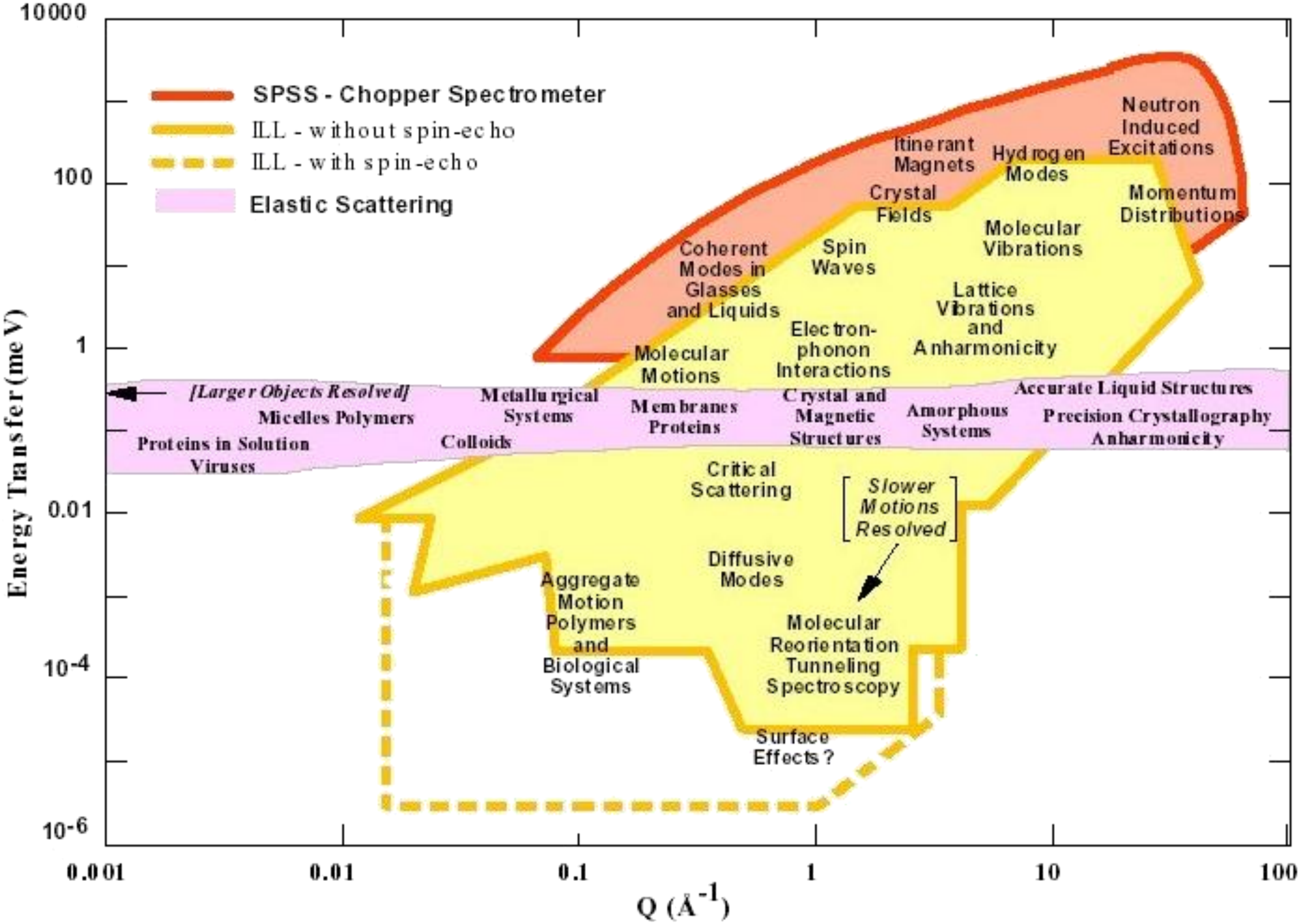
Neutrons in Condensed Matter Research



The ESS Project, Vol II, ed. By D. Richter (FZ Jülich, 2002), p.5-4

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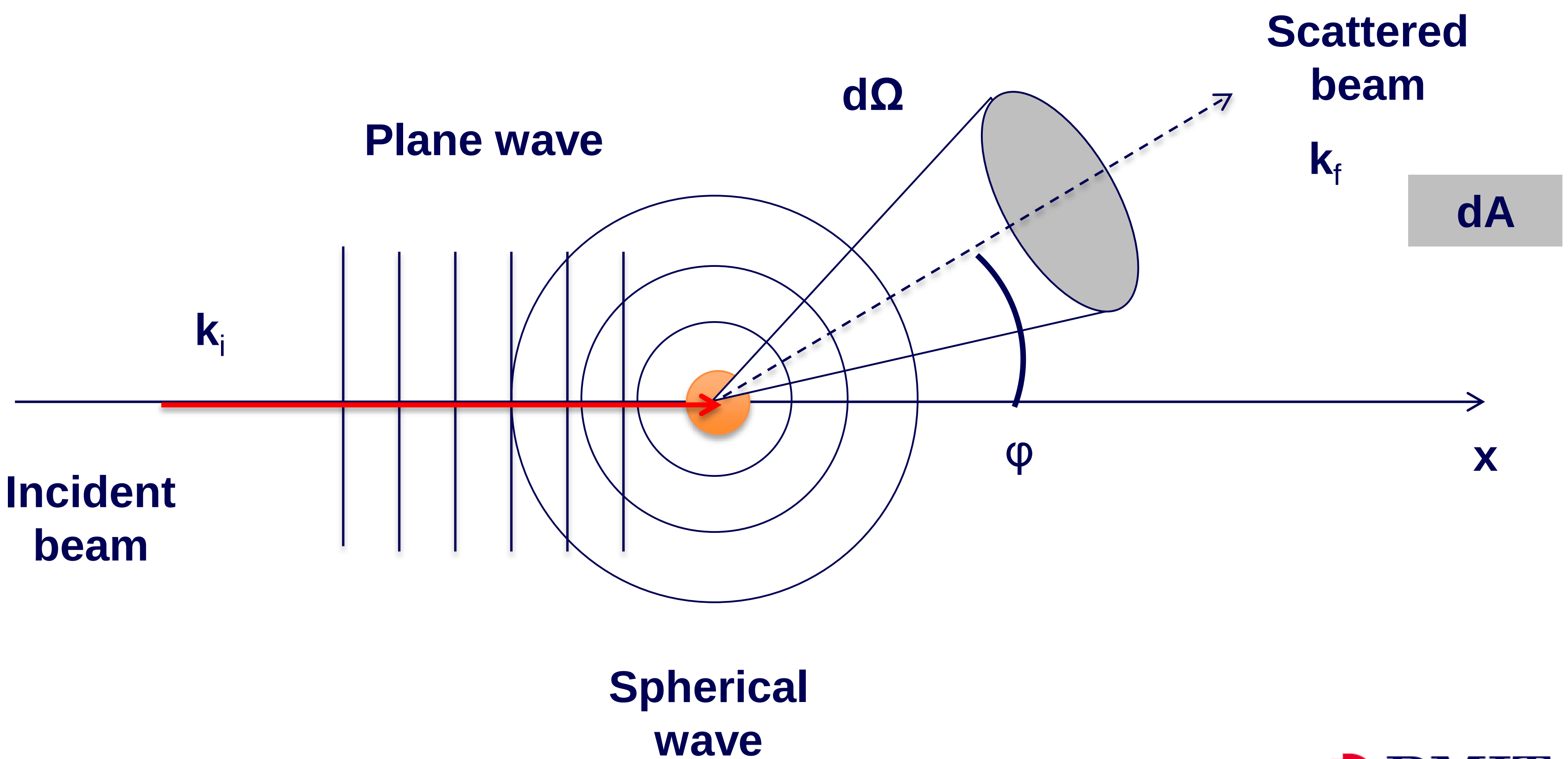
Total Cross-Section

- Scattering occurs in an elementary cone of solid angle dΩ



Total Cross-Section

- Total cross-section defined by:

$$\sigma_{tot} = \frac{no.\,of\ neutrons\ scattered\ in\ all\ directions\ per\ second}{incident\ flux\ (I_0)}$$

- Incident plane wave of neutrons: $\psi_i = e^{-ikx}$ k = wavenumber

- The probability of finding a neutron in a volume dV is: $|\psi_i|^2 dV$ however, $|\psi_i|^2 = 1$

  – $\psi_i = e^{-ikx}$ refers to density of one neutron per unit volume in all space

- The flux of neutrons incident normally on unit area per second is:

$$I_0 = neutron\ density\ \times velocity = v$$

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Total Cross-Section

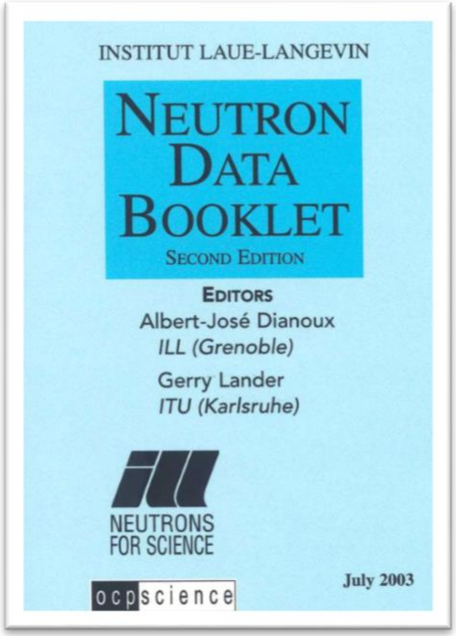
- Wave scattered by an isolated nucleus: $\psi_f = -b\frac{e^{-ikr}}{r}$

r = distance from scattering nucleus
b= scattering length of nucleus

$$=> \sigma_{tot} = \frac{I_f}{I_0} = 4\pi b^2$$

- **This is the effective area of the nucleus viewed by the neutron**
  - Units of cross-section = barns [1 barn = 10^{-28}m^2]
  - Units of scattering lengths = fermis [1 fermi = 10^{-15}m]

			b = scattering length (fermi)				σ = cross sections (barns)			
ZSymbA	p or $T_{1/2}$	I	b_c	b_+	b_-	c	σ_{coh}	σ_{inc}	σ_{scatt}	σ_{abs}
0-N-1	**10.3 MIN**	**1/2**	**-37.0(6)**	**0**	**-37.0(6)**		**43.01(2)**		**43.01(2)**	**0**
1-H			**-3.7409(11)**				**1.7568(10)**	**80.26(6)**	**82.02(6)**	**0.3326(7)**
1-H-1	99.985	1/2	-3.7423(12)	10.817(5)	-47.420(14)	+/-	1.7583(10)	80.27(6)	82.03(6)	0.3326(7)
1-H-2	0.0149	1	6.674(6)	9.53(3)	0.975(60)		5.592(7)	2.05(3)	7.64(3)	0.000519(7)
1-H-3	12.26 Y	1/2	4.792(27)	4.18(15)	6.56(37)		2.89(3)	0.14(4)	3.03(5)	< 6.0E-6
2-He			**3.26(3)**				**1.34(2)**	**0**	**1.34(2)**	**0.00747(1)**
2-He-3	0.00013	1/2	5.74(7)	4.374(70)	9.835(77)	E	4.42(10)	1.532(20)	6.0(4)	5333.0(7.0)
2-He-4	0.99987	0	3.26(3)				1.34(2)	0	1.34(2)	0
3-Li			**-1.90(3)**				**0.454(10)**	**0.92(3)**	**1.37(3)**	**70.5(3)**
3-Li-6	7.5	1	2.0(1)	0.67(14)	4.67(17)	+/-	0.51(5)	0.46(5)	0.97(7)	940.0(4.0)



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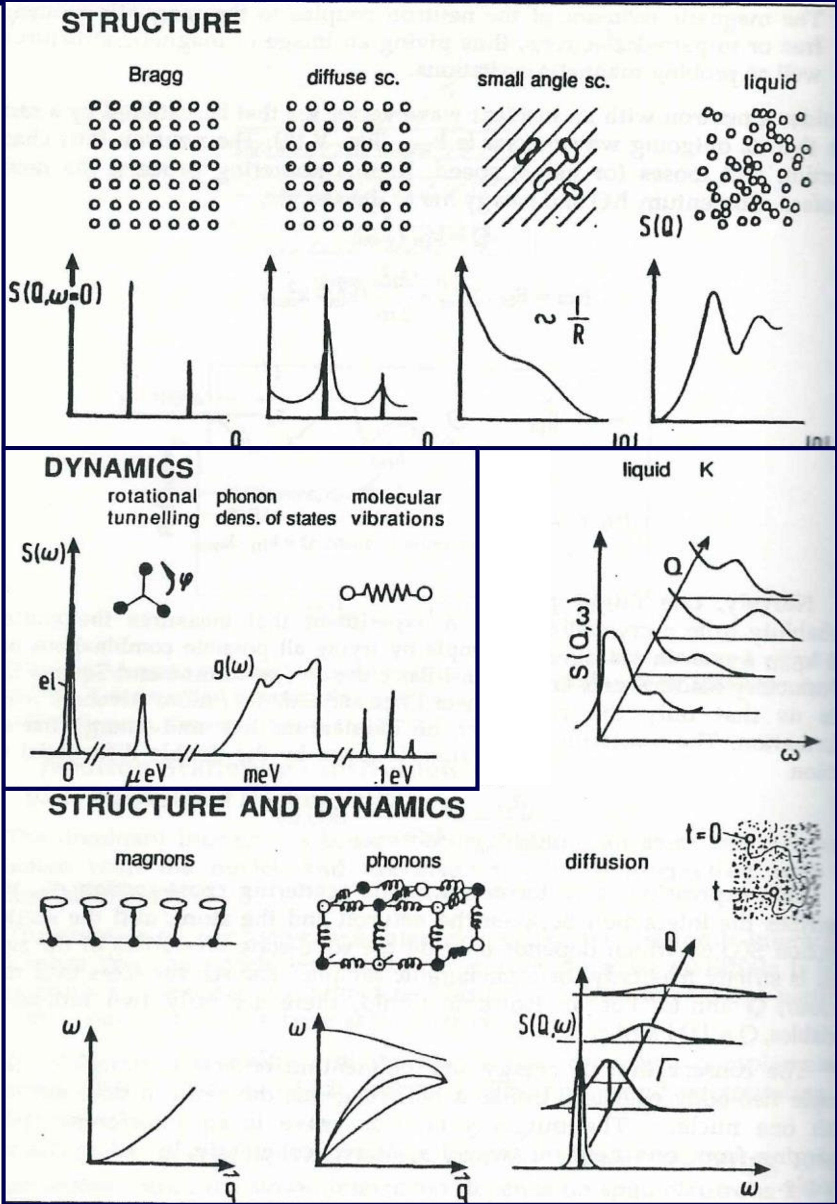
Scattering Function

- Scattering per atom is given by a double differential cross section
  - Scattering cross section σ_s
  - Scattering function $S(Q,\omega)$

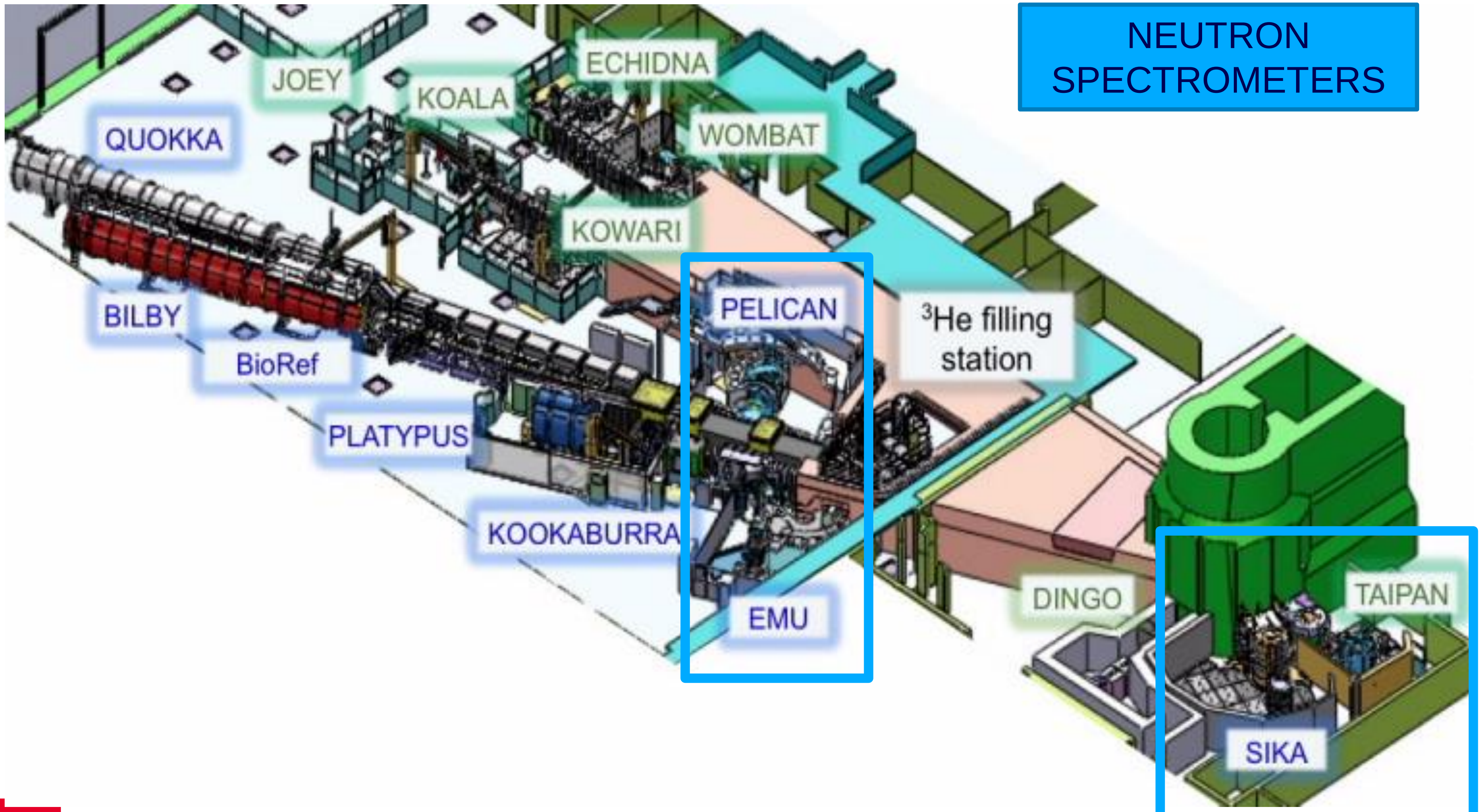
- Elastic scattering: $\omega=0$
- Inelastic scattering at any Q
  - Localised motion
- Q-dependent frequencies in $S(Q,\omega)$
  - Propagating motions in r(t)



Neutron and Synchrotron Radiation for Condensed Matter Studies, J. Baruchel, 1993

2. Inelastic neutron scattering

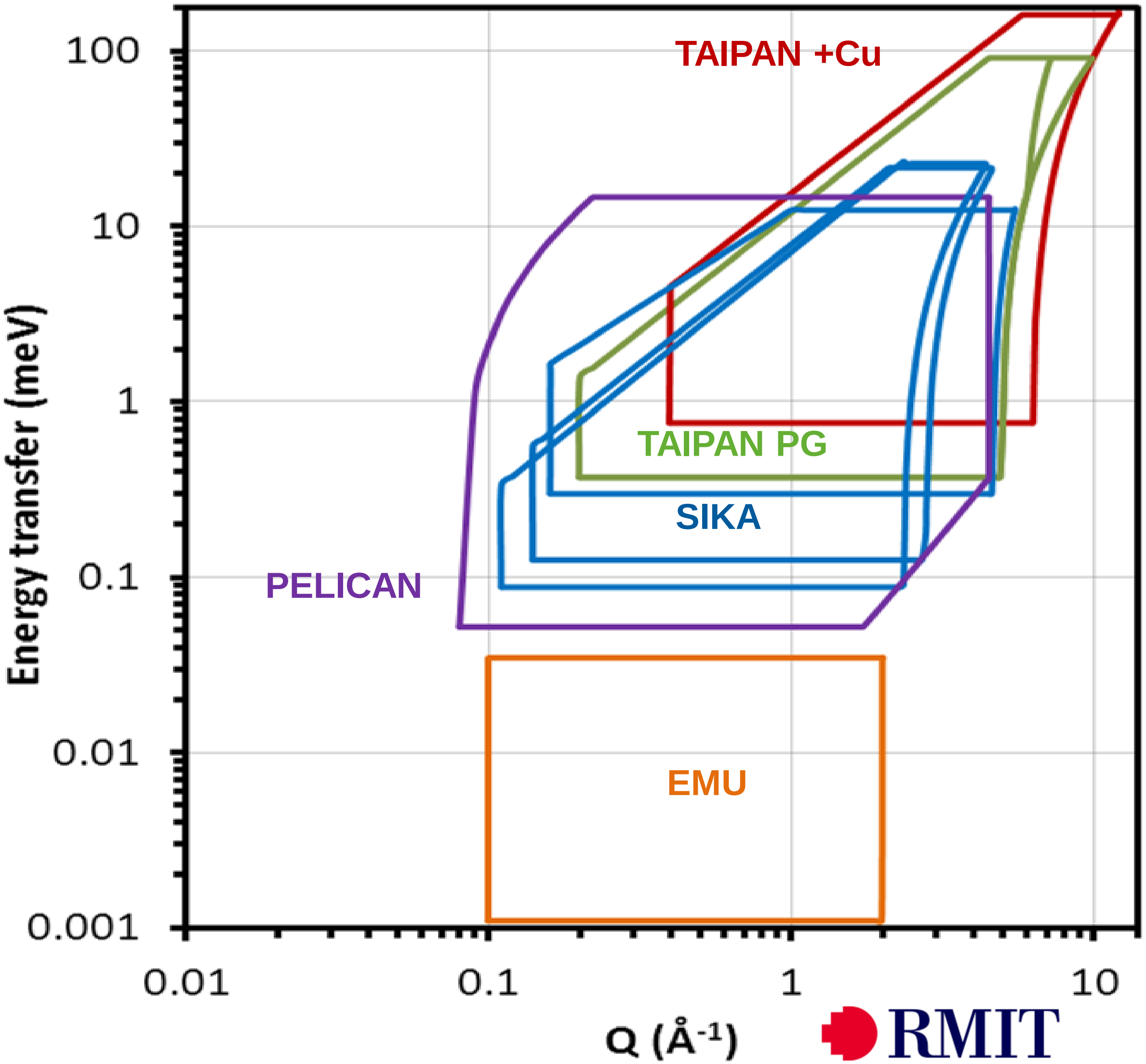
Neutron spectrometers at ANSTO



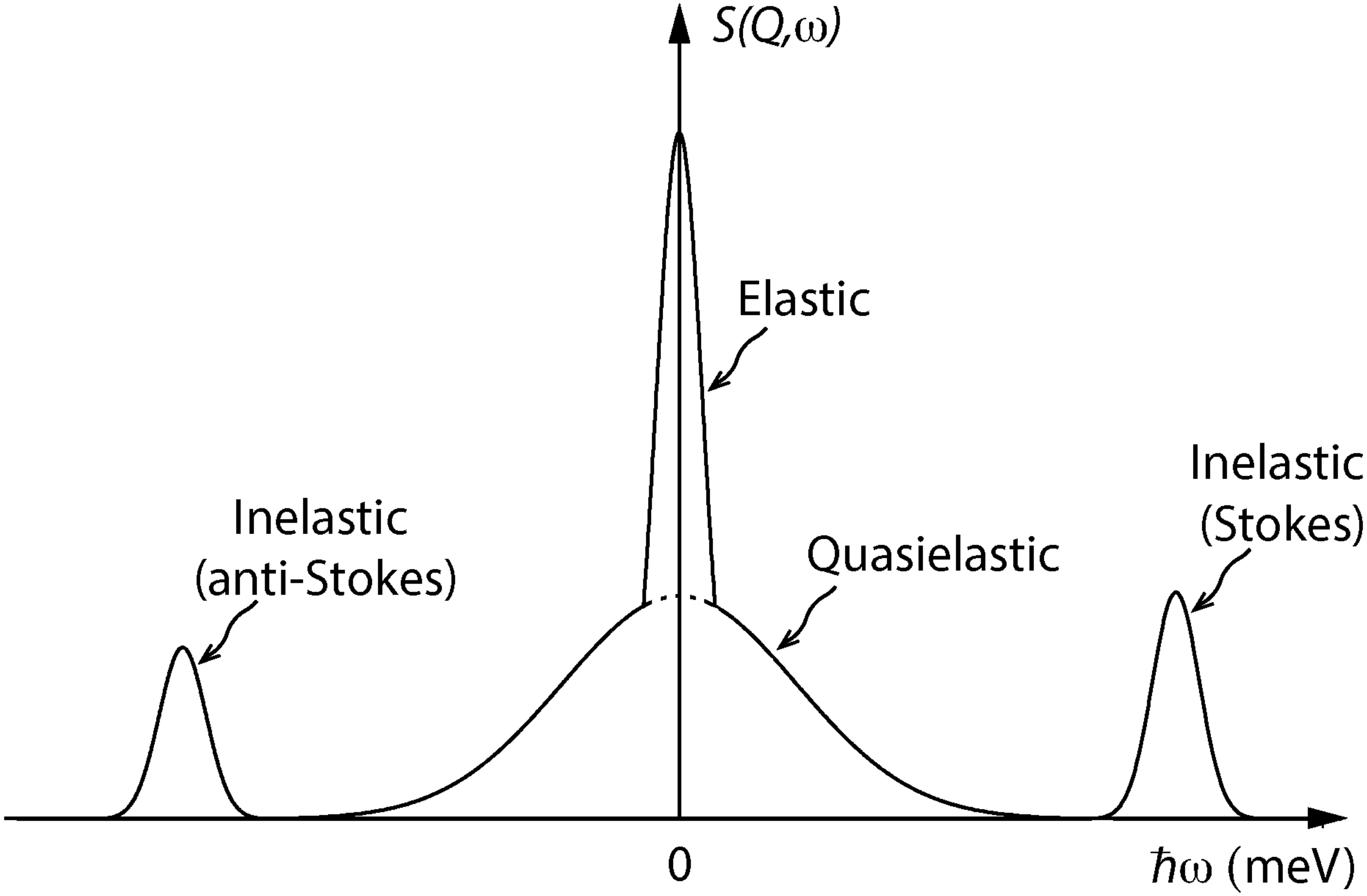
Energy resolution of ACNS spectrometers

- Capabilities for Dynamics and Excitations at OPAL

F. Klose, P. Constantine, S.J. Kennedy and R.A. Robinson. *J. Phys.: Conf. Ser.* ***528*** *(2014) 012026*



Inelastic Data Traces

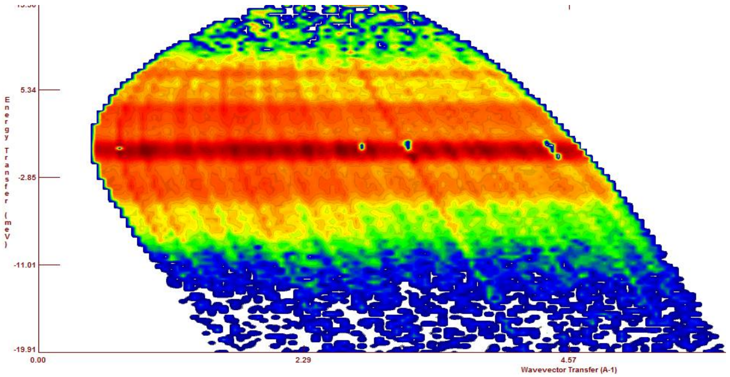


Maths Karlsson. Phys. Chem. Chem. Phys., 2015,17, 26-38

Kinematics of inelastic scattering

- Remember this equation?

$$Q^2 = k_i^2 + k_f^2 - 2k_i k_f \cos 2\theta$$



- Written in terms of energy:

$$\frac{\hbar^2 Q^2}{2m_n} = E_i + E_f - 2\sqrt{(E_i E_f)}\cos 2\theta$$

$$= 2E_i - \hbar\omega - 2\sqrt{E_i(E_i - \hbar\omega)}\cos 2\theta$$

For direct geometry spectrometer

$$= 2E_f + \hbar\omega - 2\sqrt{E_i(E_f + \hbar\omega)}\cos 2\theta$$

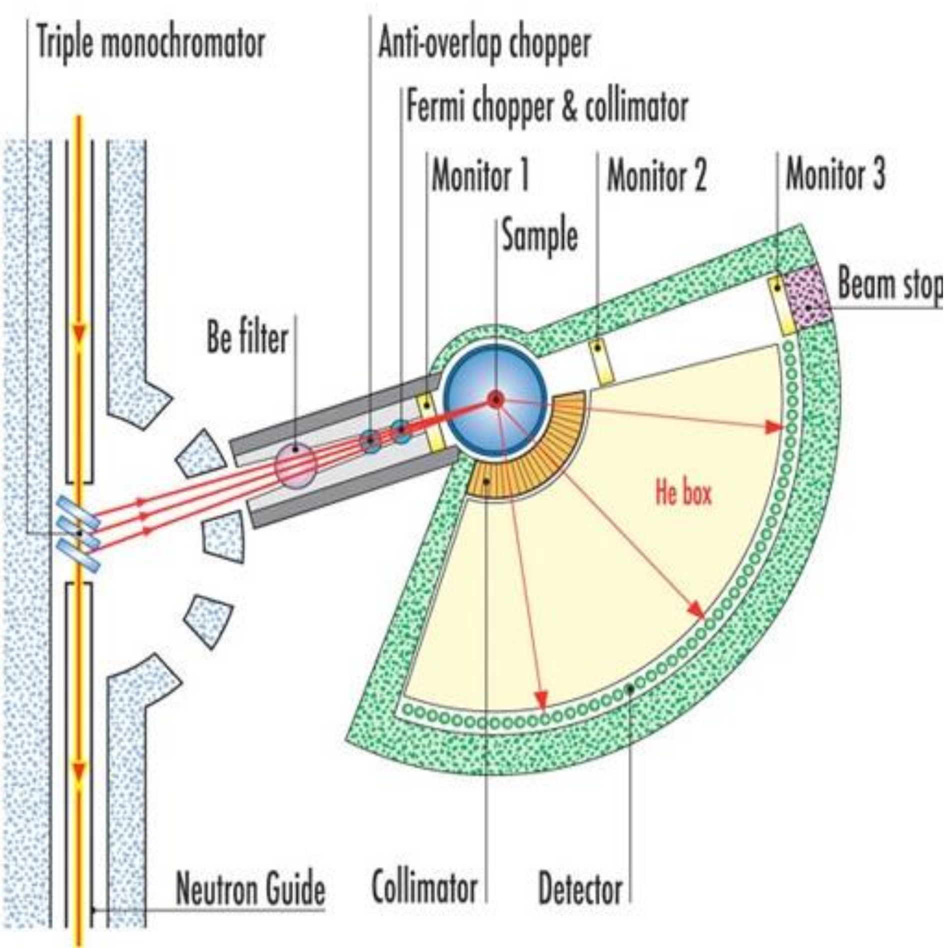
For indirect geometry spectrometer

3. Time-of-flight neutron spectroscopy

Time-of-flight Spectrometer

- Time-of-flight spectrometer (TOF)
  - – Monochromator
    - Selects neutron wavelength
  - – Choppers
    - Define E_i
  - – Sample
    - Scatters neutrons
  - – Detectors
    - Register time of arrival of neutrons -> E_f obtained

Direct geometry spectrometer



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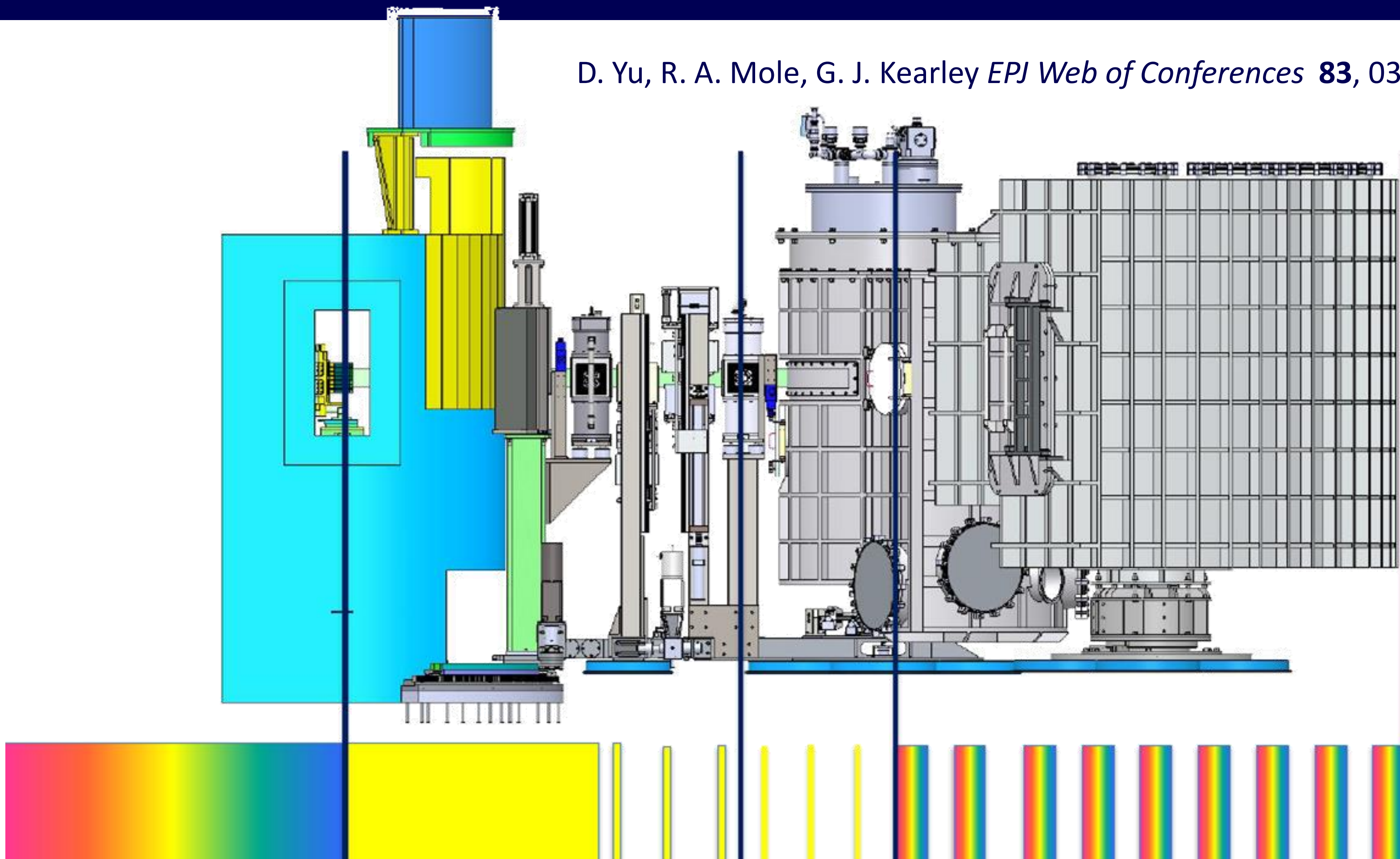
D. Yu, R. A. Mole, G. J. Kearley *EPJ Web of Conferences* **83**, 03019 (2015)



Continuous spectrum of neutron energies	Monochromator selects single energy / wavelength	Fermi chopper pulses the beam	Second chopper refines the beam	Neutrons scattered by the sample are time shifted.	Scattered neutrons carry both time and position information to determine E_f.

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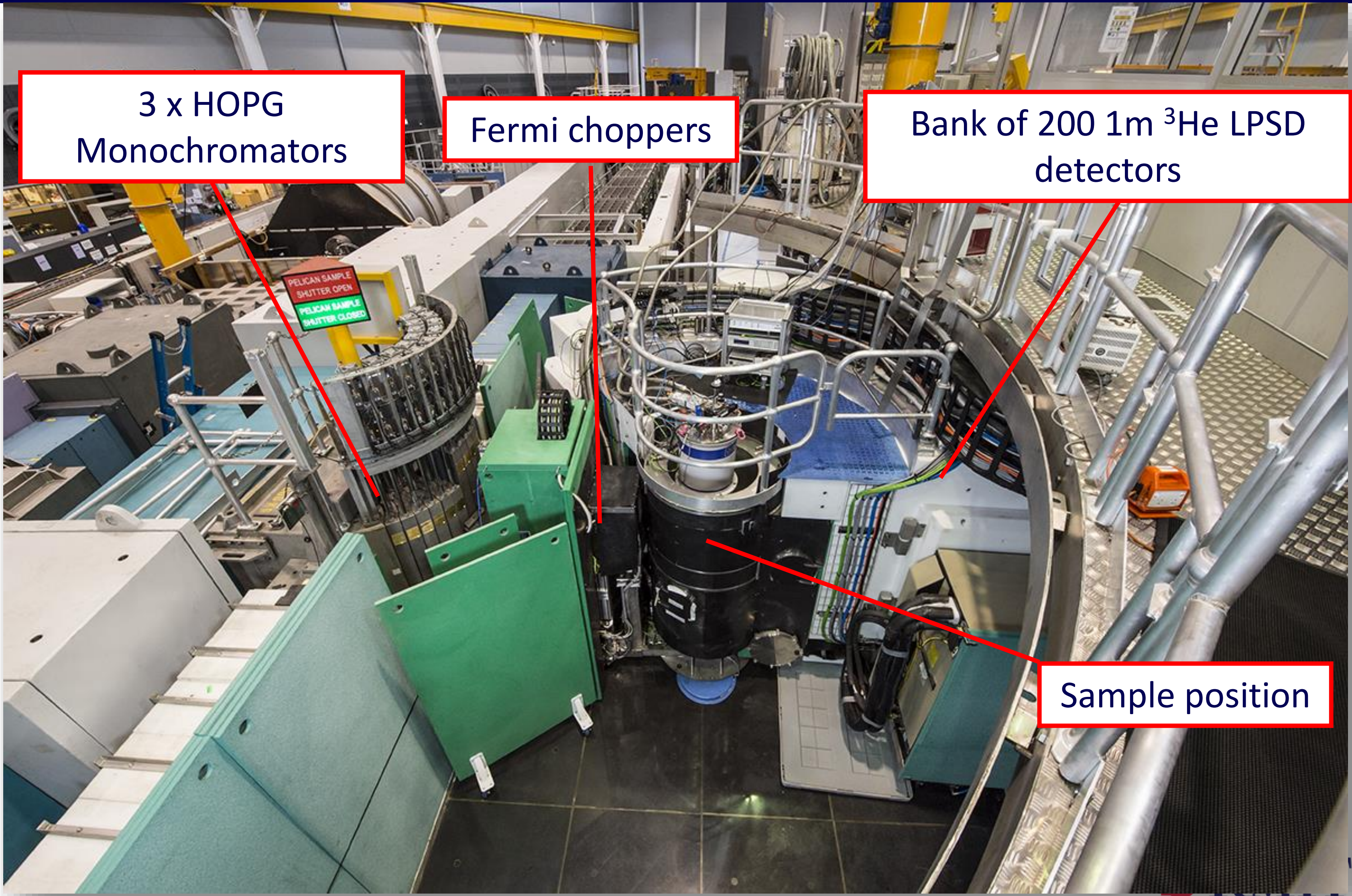
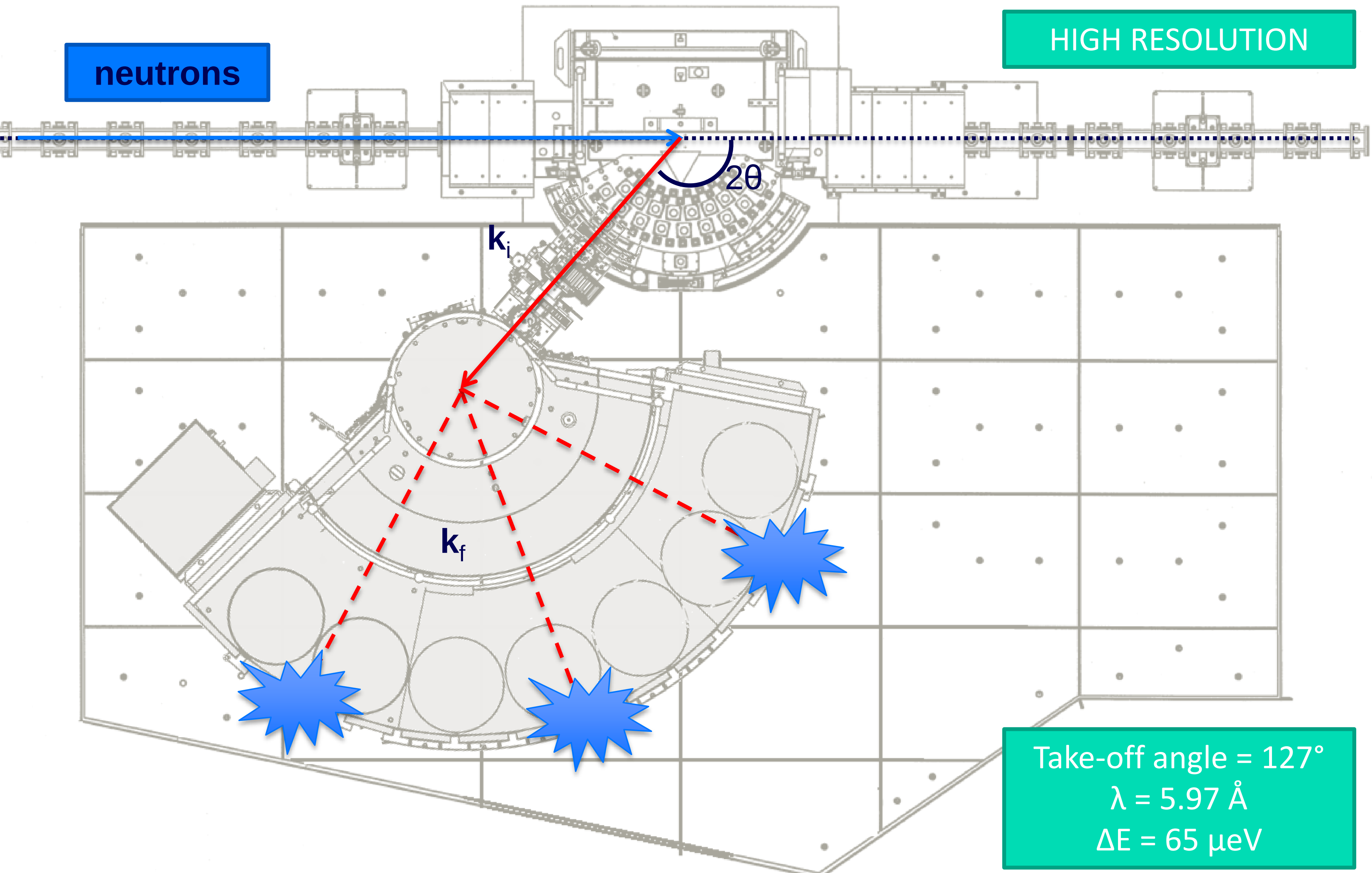
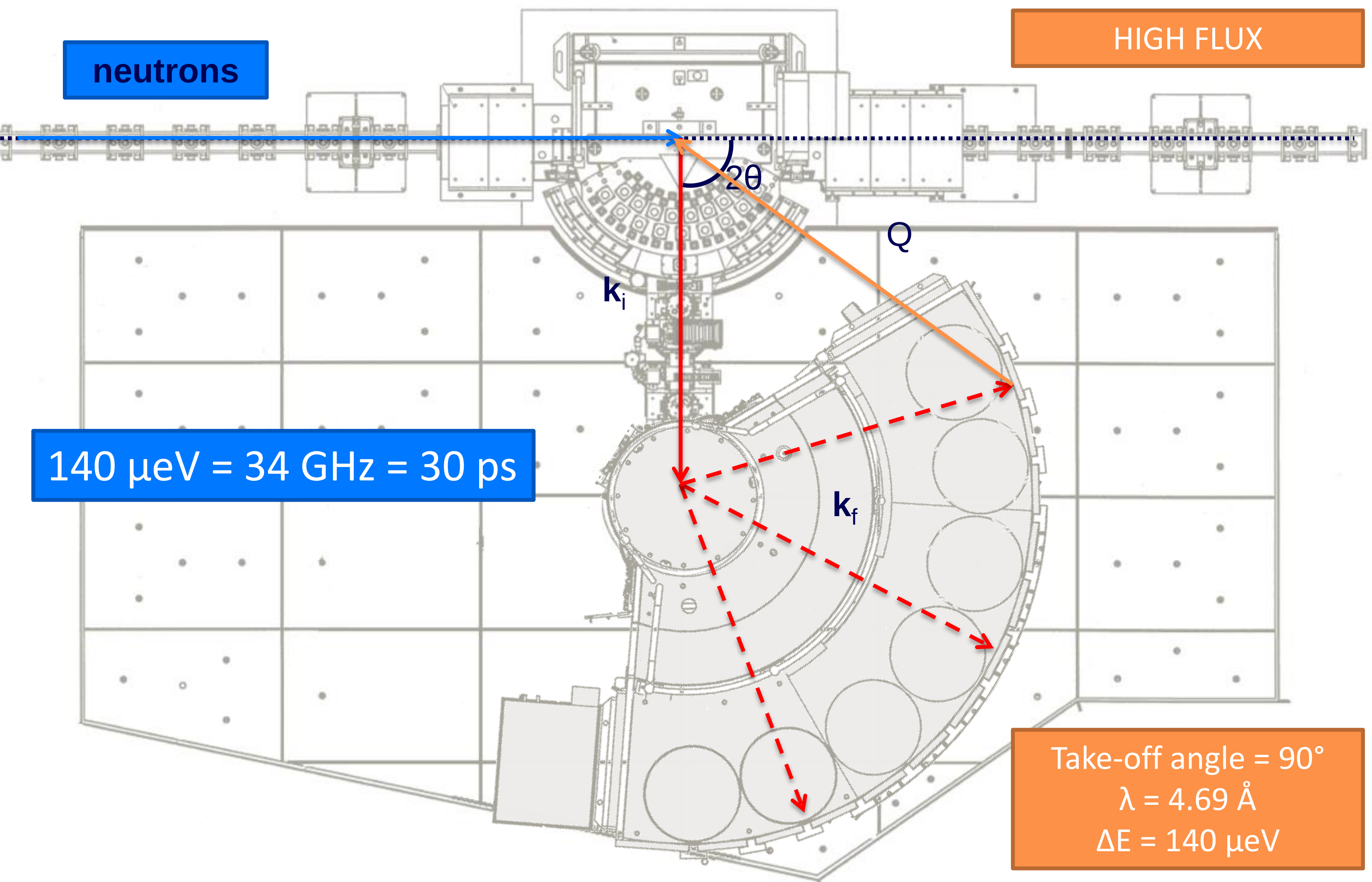

3 x HOPG Monochromators
Fermi choppers
Bank of 200 1m ^{3}He LPSD detectors
Sample position

PELICAN – Wavelength options

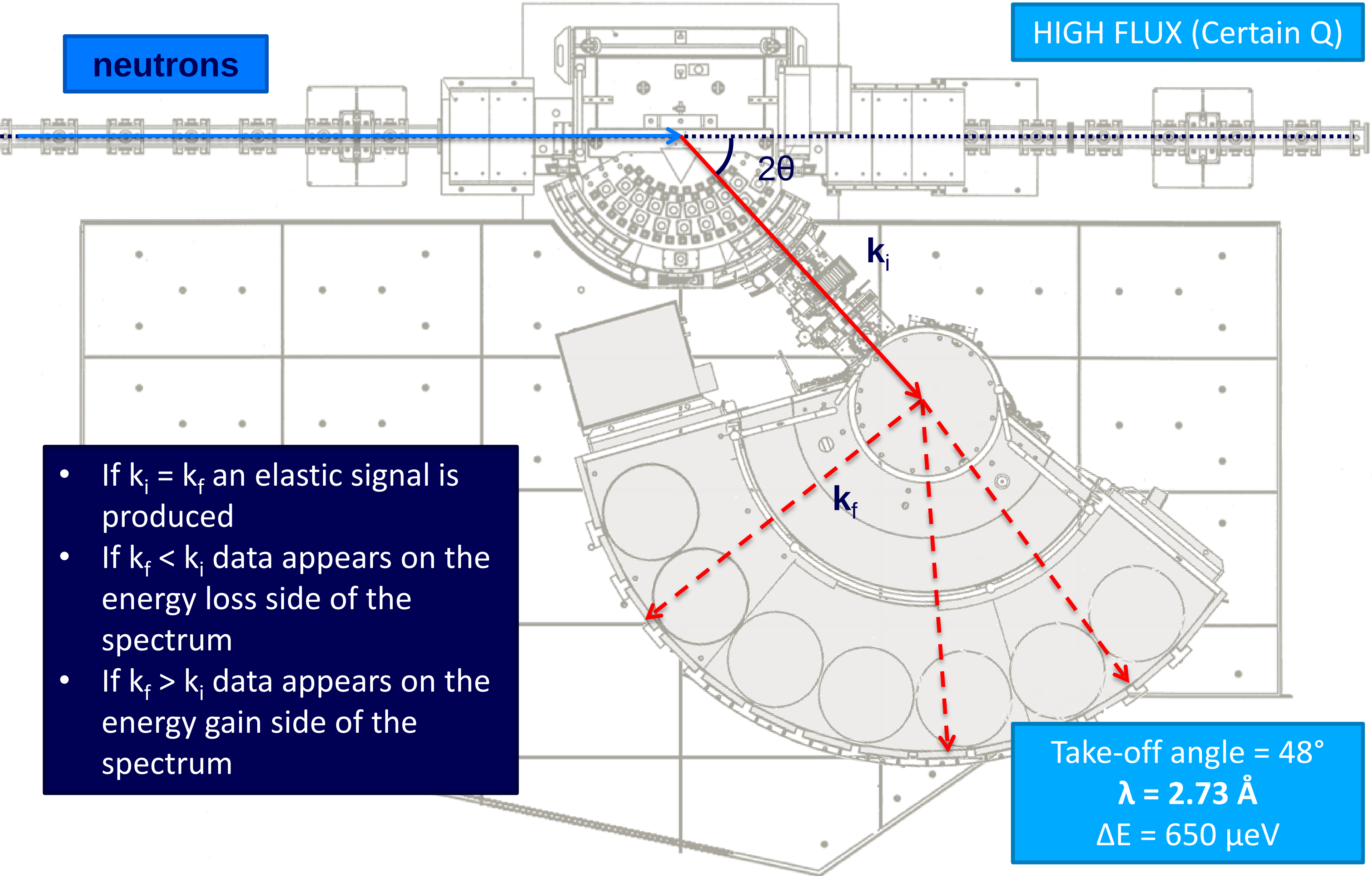


Take-off angle = 127°
λ = 5.97 Å
ΔE = 65 μeV

PELICAN – Wavelength options



PELICAN – Wavelength options



HIGH FLUX (Certain Q)

- If $k_i = k_f$ an elastic signal is produced
- If $k_f < k_i$ data appears on the energy loss side of the spectrum
- If $k_f > k_i$ data appears on the energy gain side of the spectrum

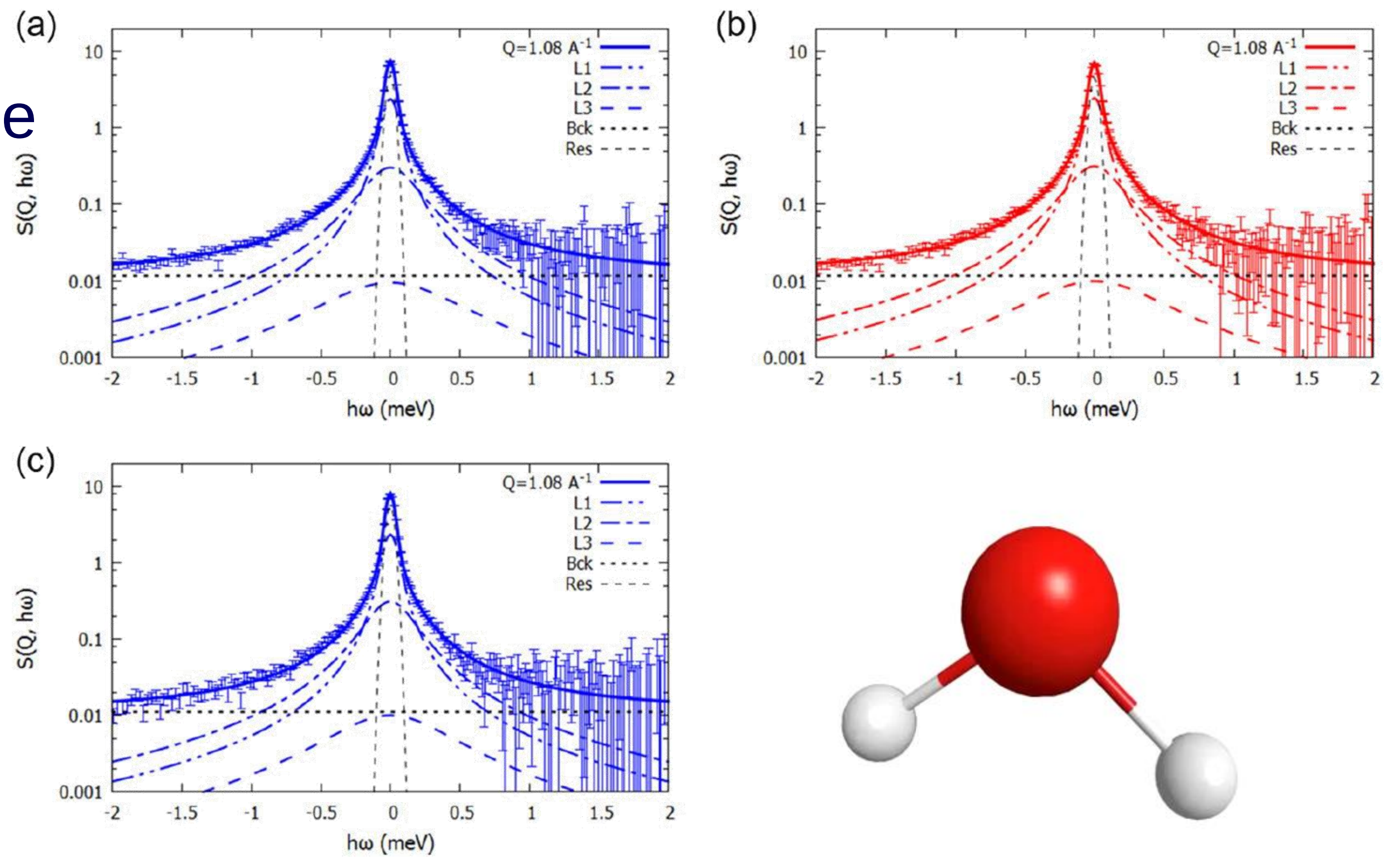
Take-off angle = 48°
λ = 2.73 Å
ΔE = 650 μeV

Data from PELICAN

Water desorption and absorption in sodium montmorillonite

QENS

- Analysis of the shape and width of the quasi-elastic peak reveals dynamics information
- Direct correlation between energy and frequency of motion



Gates et al. Applied Clay Science **147** (2017)

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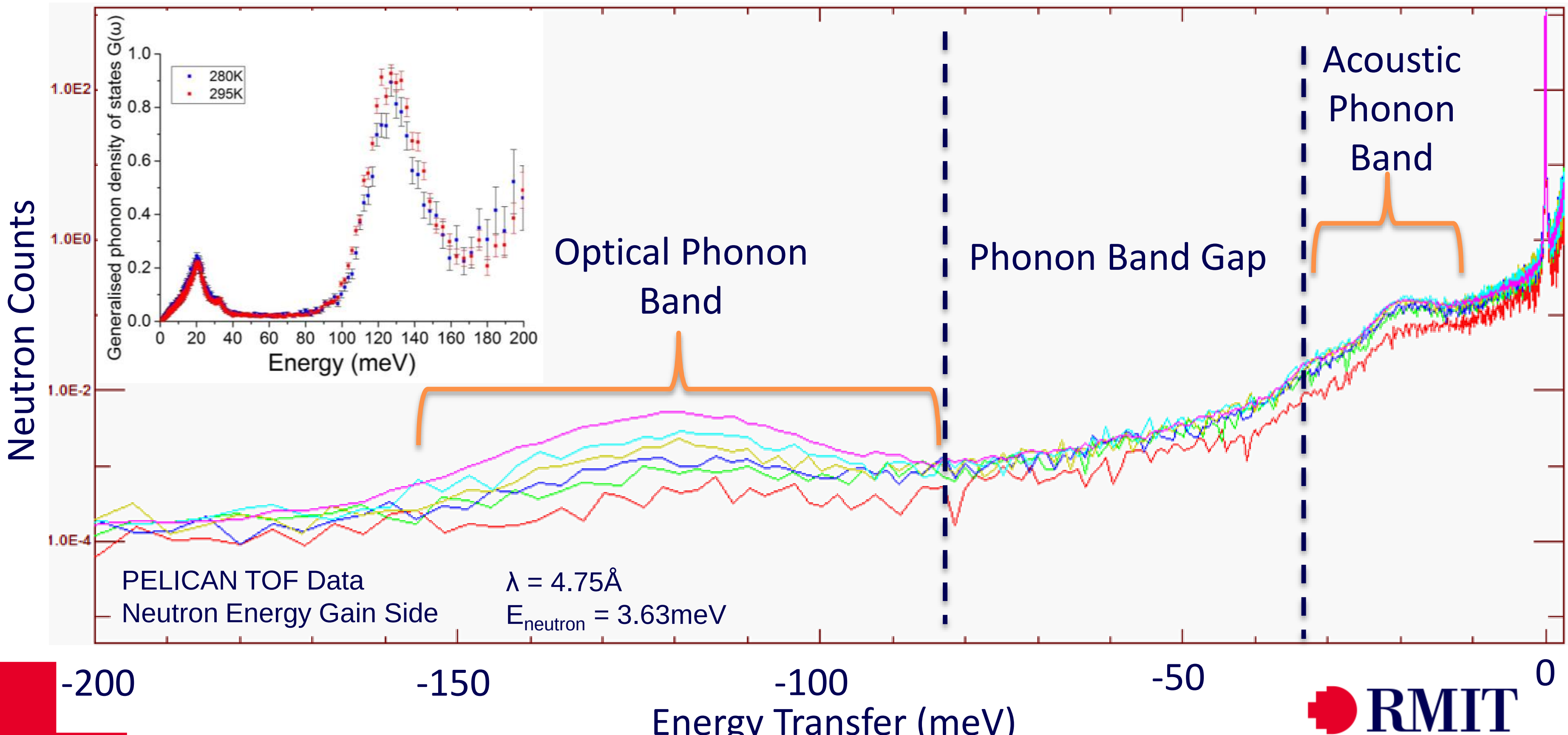
Science on PELICAN: Case study 1

- Vibrational density of states of crystalline and amorphous solids

Wang et al. Jpn. J. Appl. Phys. **56** (2017)



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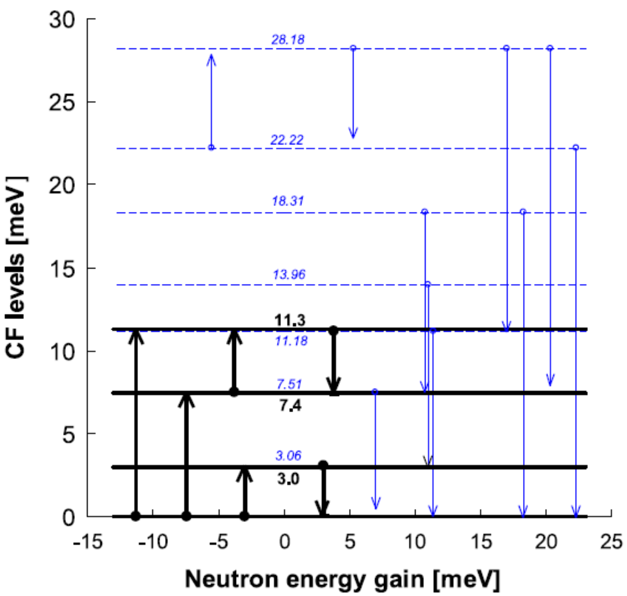
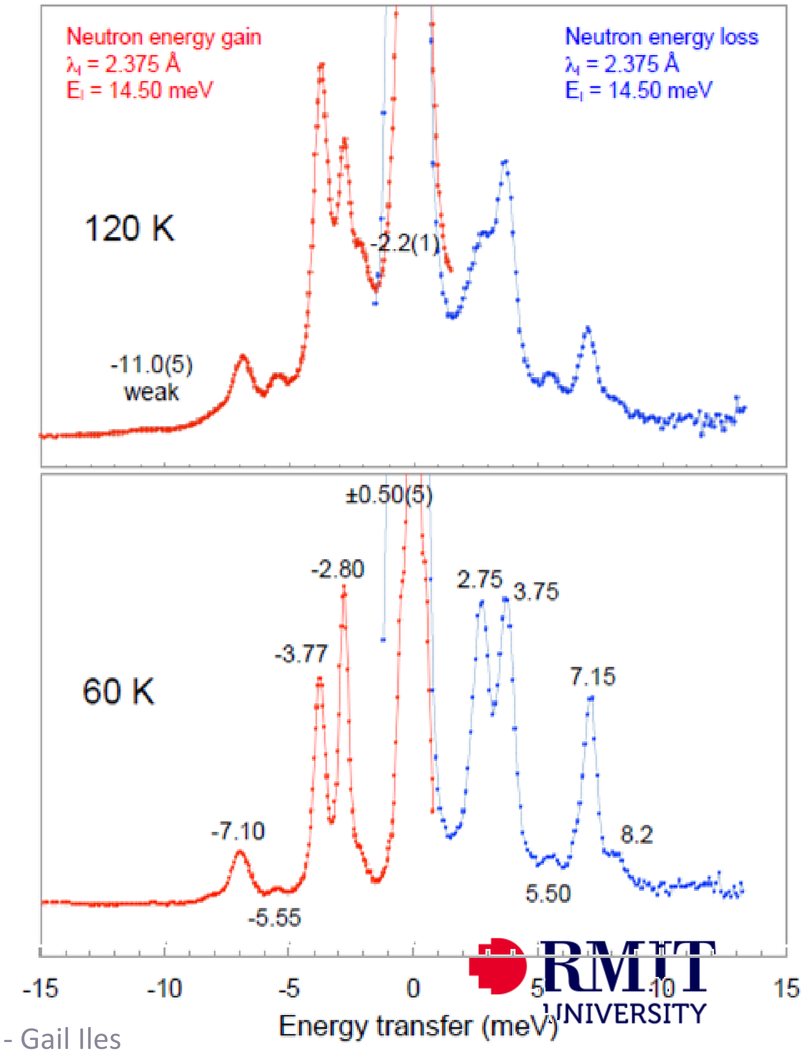
Science on PELICAN: Case study 2

- Properties of crystal-field splittings
- The Er^{3+} (J = 15/2) CF scheme has the relatively large number of eight Kramers doublets.



Stewart et al. (In preparation)

Crystal field interpretation of bulk magnetic behavior in **$ErNiAl_4$**



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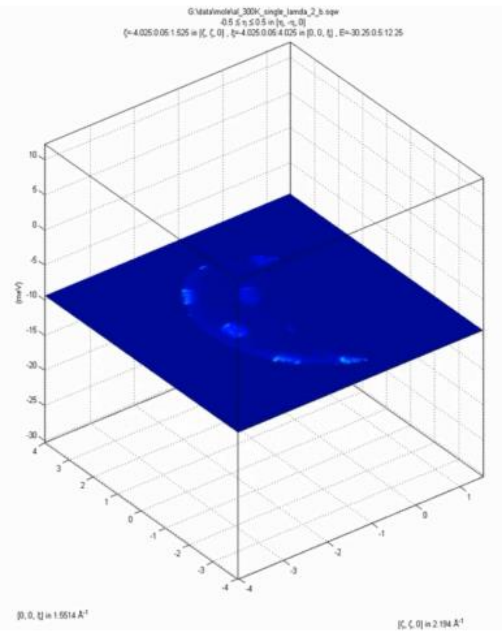
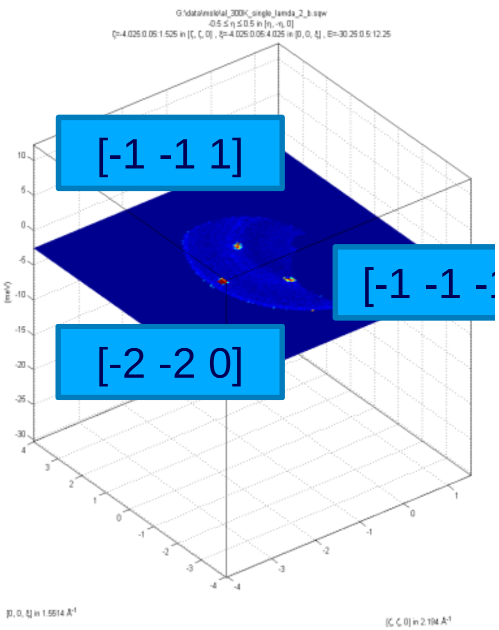
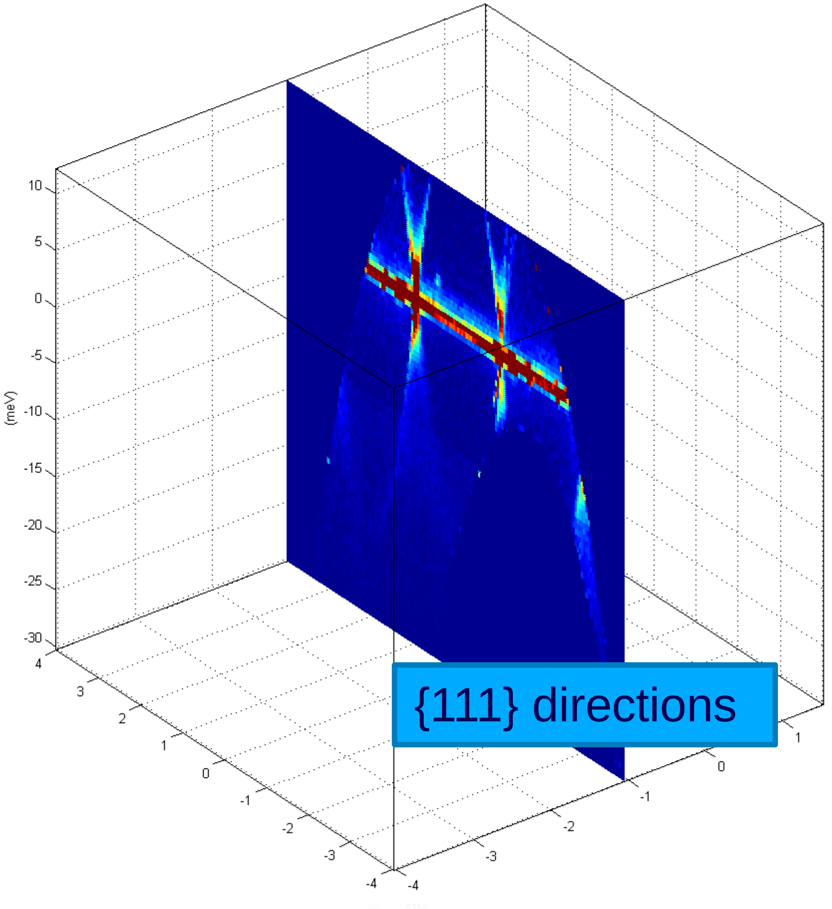
Science on PELICAN: Case study 3

- Single-crystal samples

Aluminium single crystal

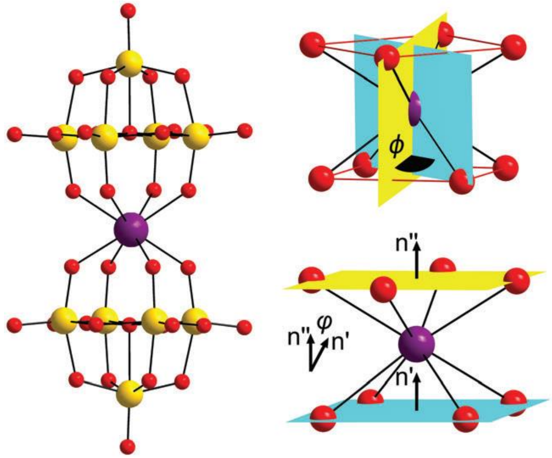






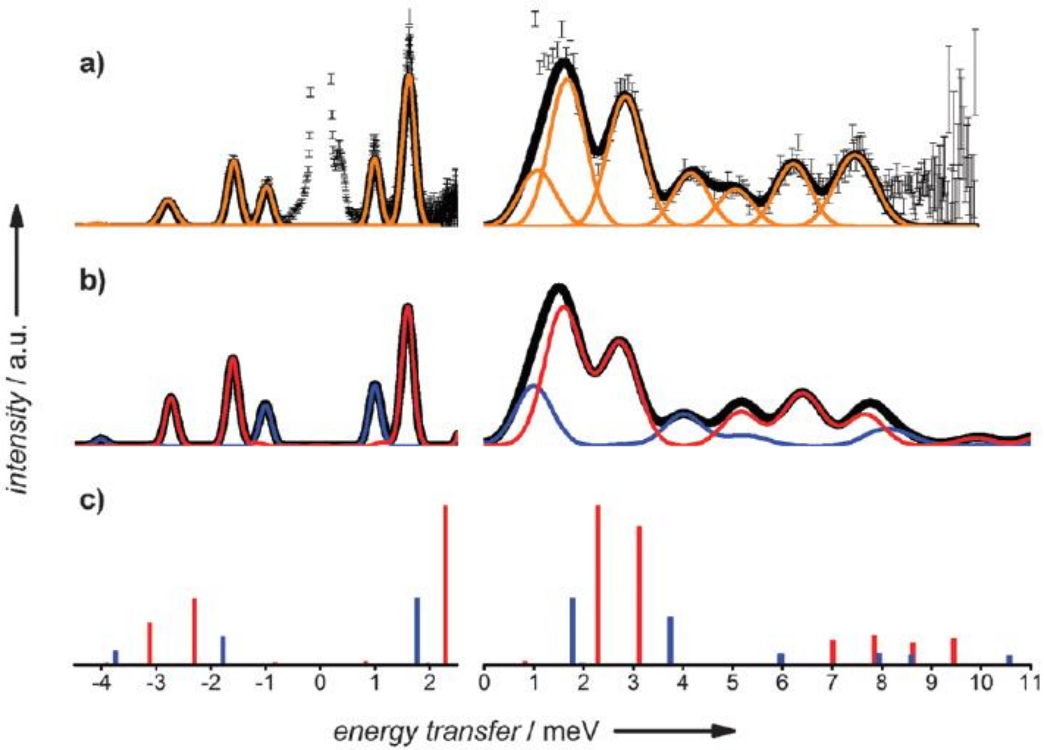
Science on PELICAN: Case study 4

- Single-molecule magnets
  - Tiny rotation of the dihedral angle gives a 1 meV shift = a 10 K change in thermal energy



Molecular structure (left) and representations of the two distortion angles of the Tb coordination (right) for the [Tb(W5O18)2]9 polyanion in Tb; atom colour code: W (yellow), O (red) and Tb (violet).

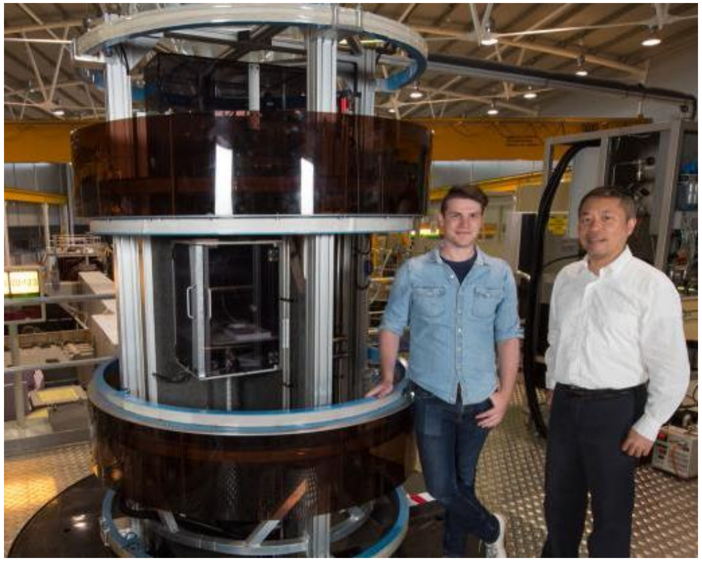
INS spectra of Tb^{D} at λ= 4.74 (left) and λ = 2.37 Å (right) at 30 K.



Vonci et al. Chem. Commun. (2015)

PELICAN Capabilities

- Gas-loading
- Humidity variation
- Polarised neutrons
- Magnetic field
- Dilution temperatures



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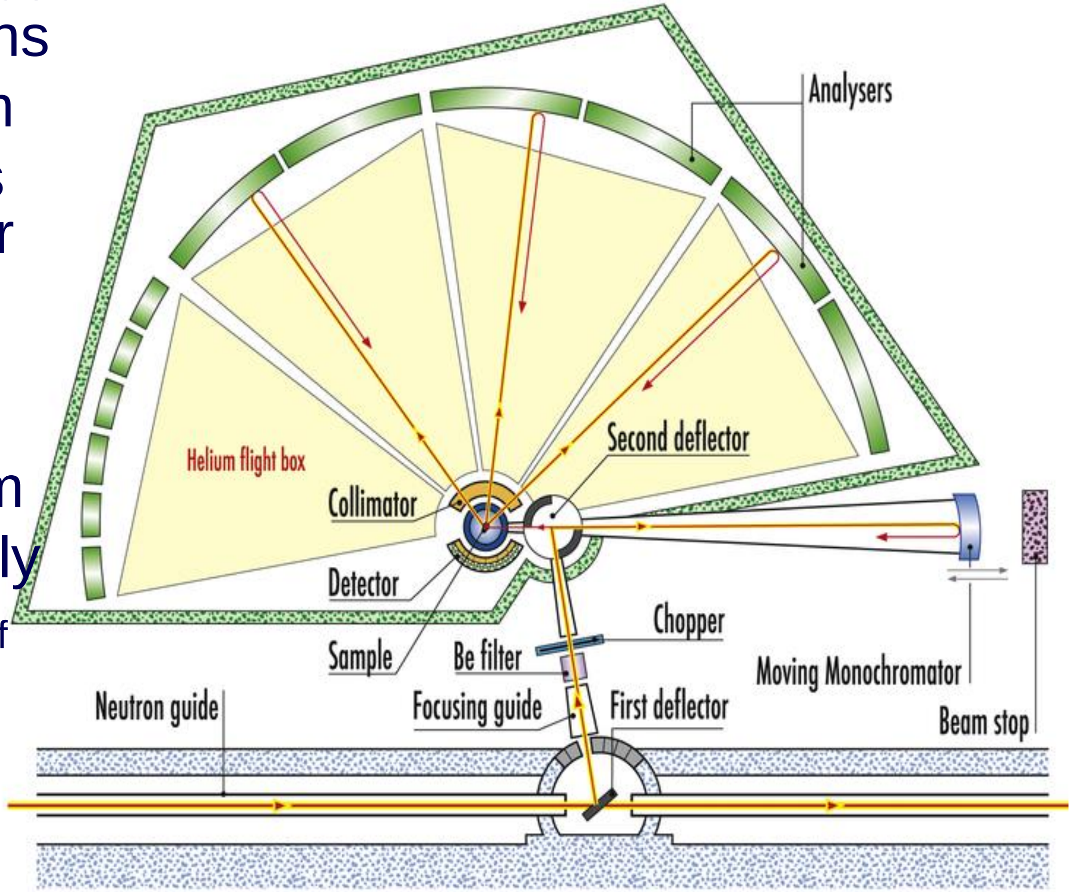
4. Backscattering Spectroscopy

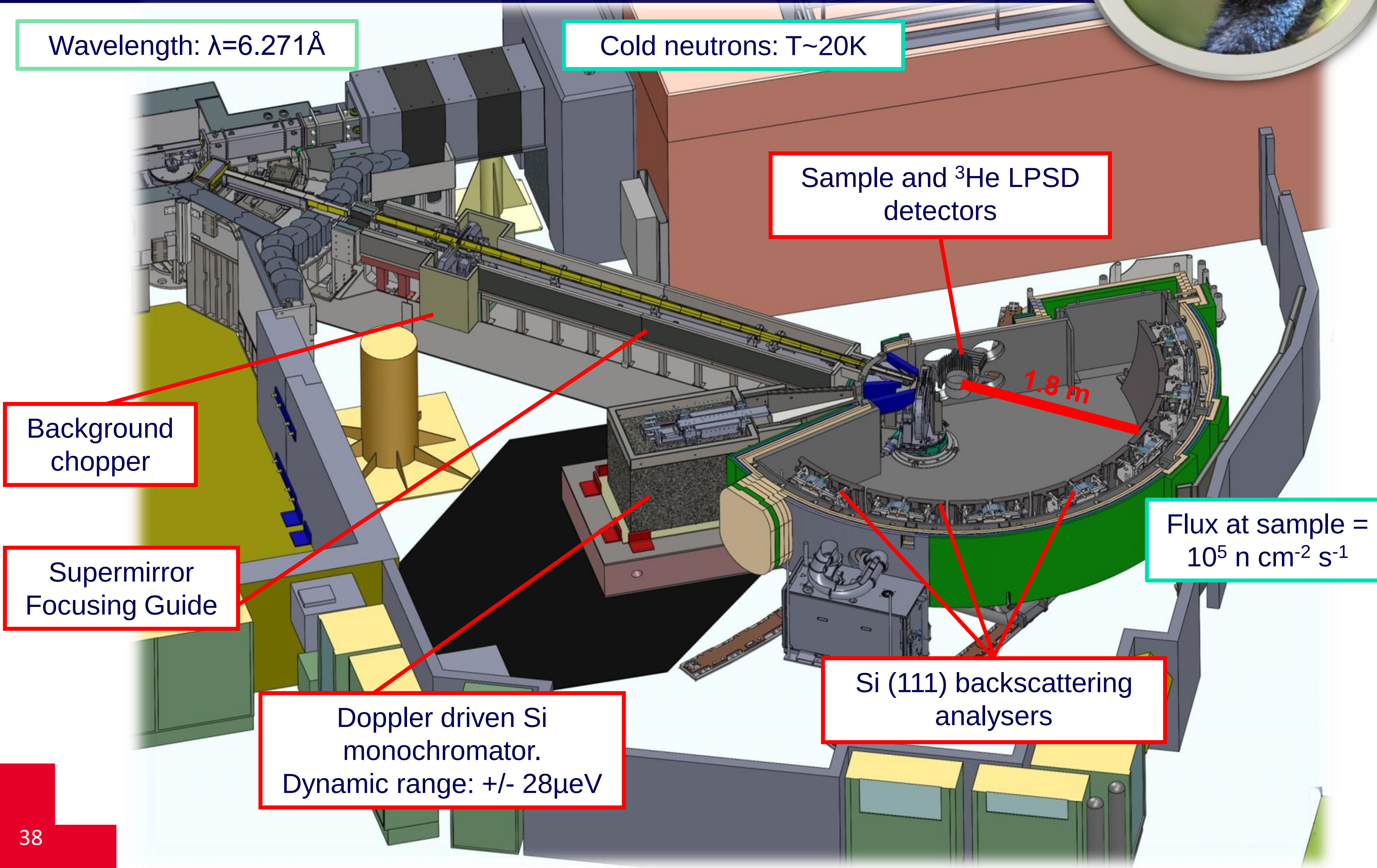
Backscattering Spectrometer

- Premonochromator – defines initial wavelength of neutrons
- Chopper – pulses the beam
- Deflecting chopper – sends neutrons to monochromator
- Doppler-driven monochromator – varies E_i (indirect geometry)
- Sample – scatters the beam
- Analysers – backscatter only neutrons with certain $E = E_f$
- Detectors – only detect neutrons reflected by the analysers

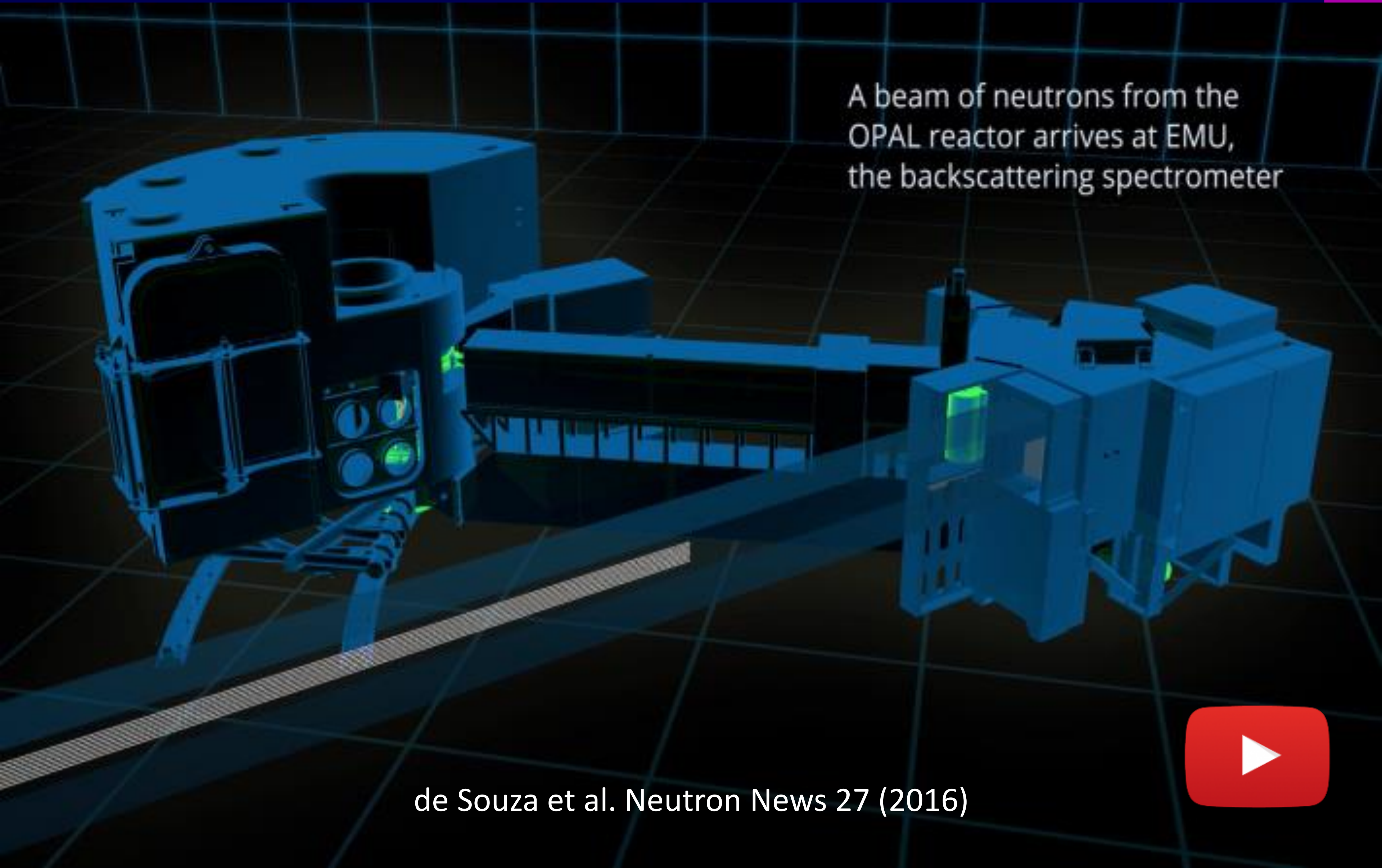


Indirect geometry spectrometer

EMU

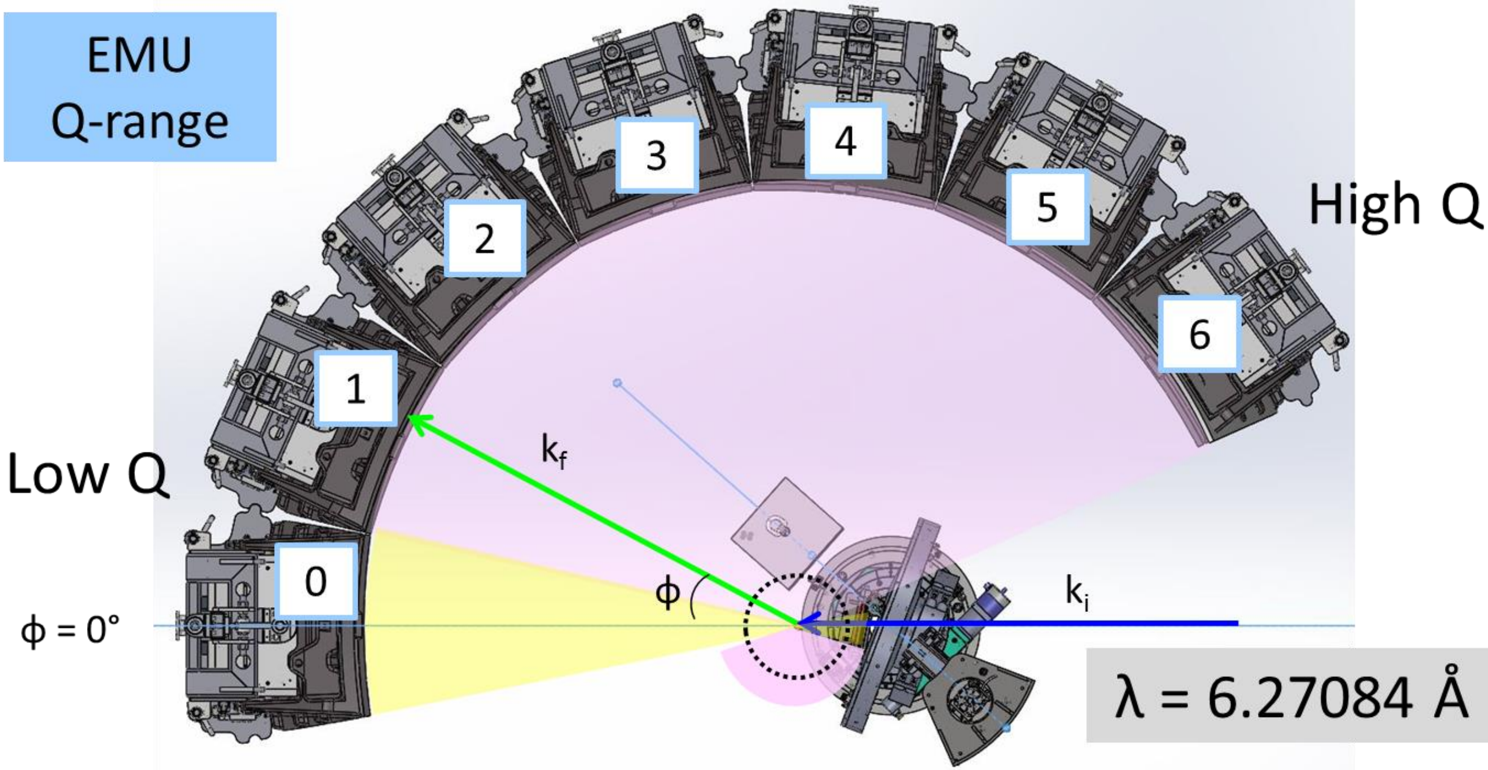

Wavelength: λ=6.271Å
Cold neutrons: T~20K
Sample and ³He LPSD detectors
1.8 m
Background chopper
Supermirror Focusing Guide
Flux at sample = 10^5 n cm^{-2} s^{-1}
Doppler driven Si monochromator. Dynamic range: +/- 28μeV
Si (111) backscattering analysers

EMU



de Souza et al. Neutron News 27 (2016)

EMU



$$Q^2 = k_i^2 + k_f^2 - 2k_ik_f\cos\phi$$

$$k = 2\pi / \lambda \sim 1.001969$$

Vertical detectors: $12° < \phi < 155°$ => 0.21 Å$^{-1} < Q < 1.96$ Å^{-1}

Horizontal detectors: $0° < \phi < 12°$ => 0.01 Å$^{-1} < Q < 0.20$ Å^{-1}

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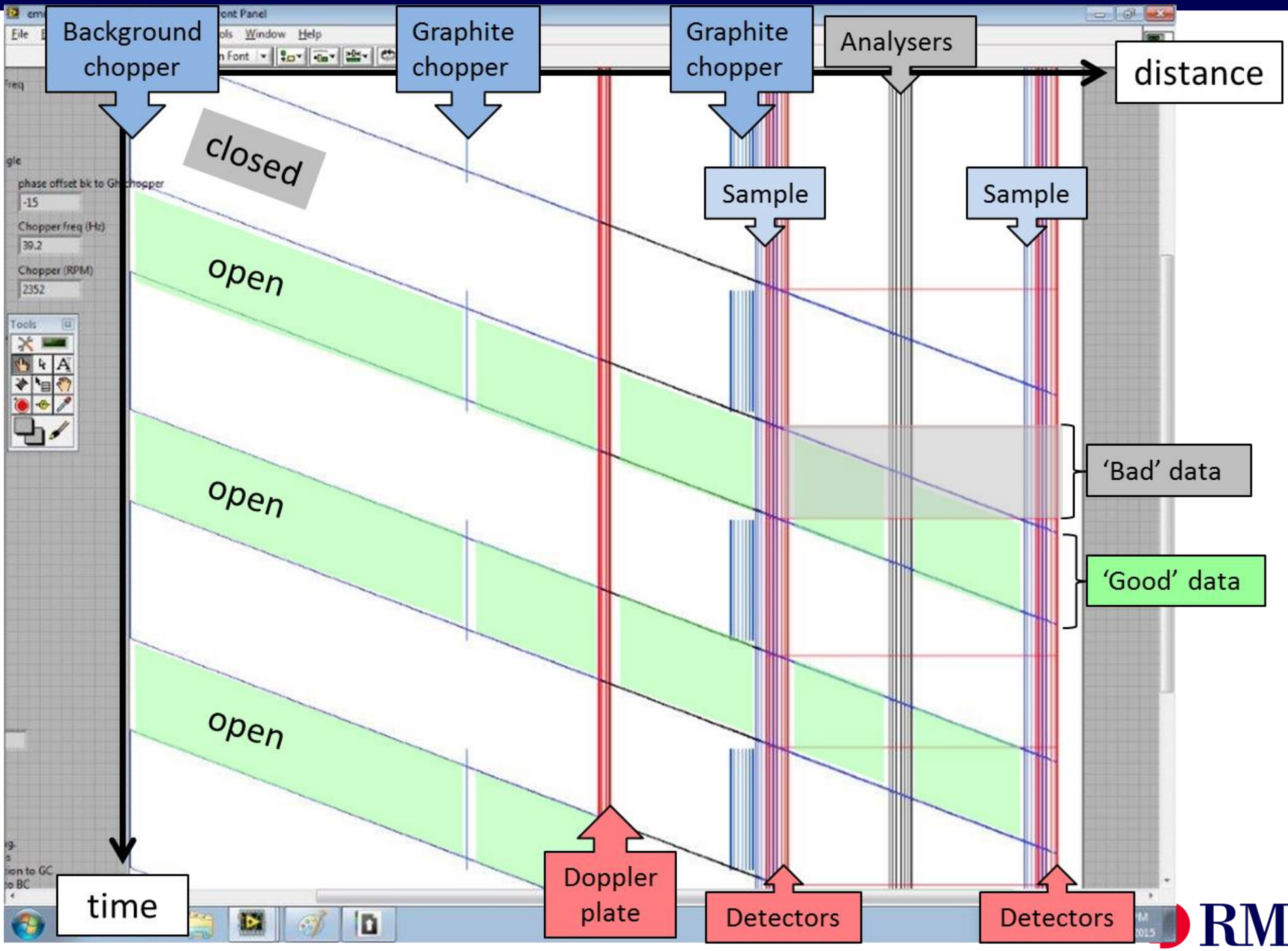
EMU Distance/Time Diagram



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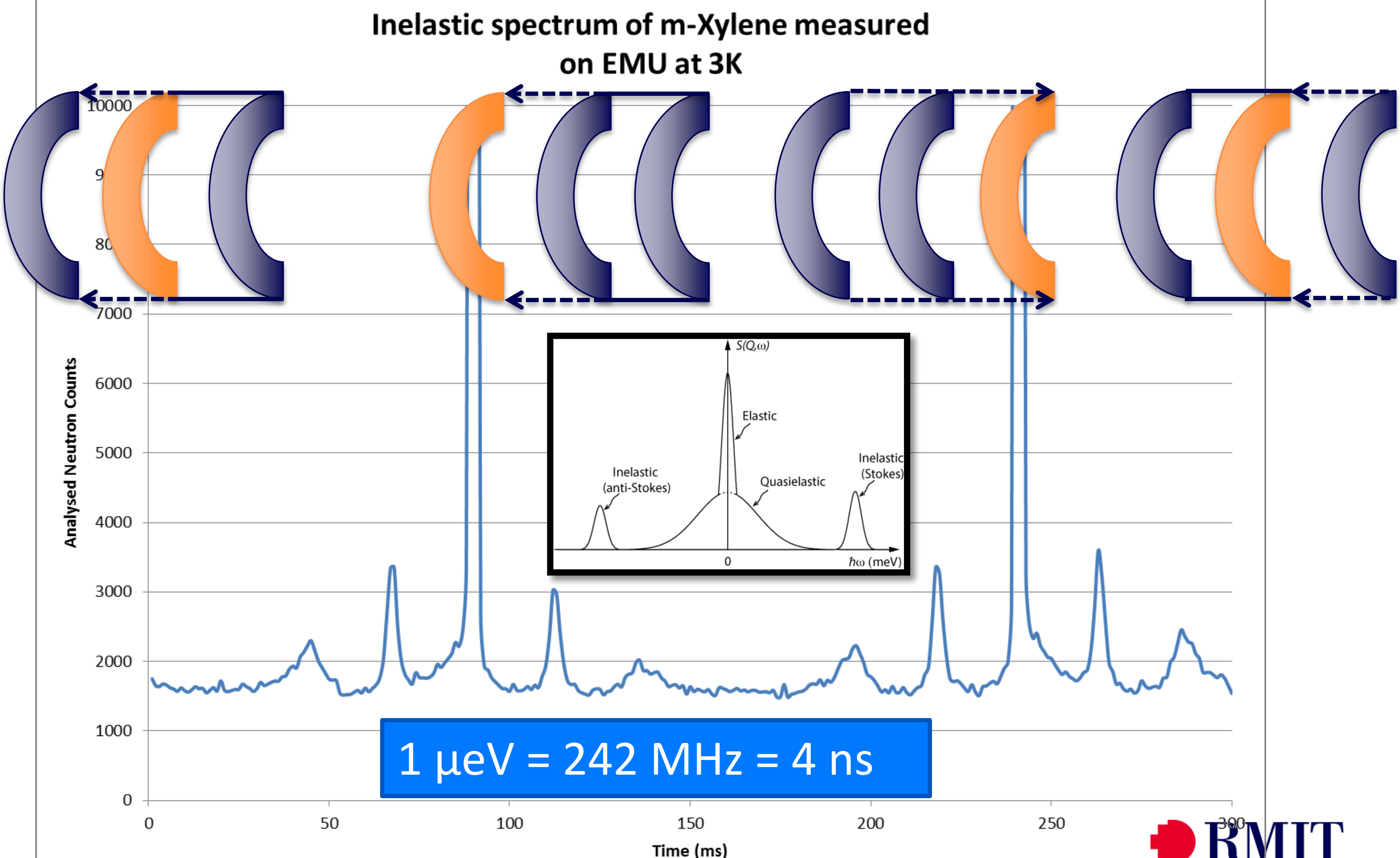
Data from EMU



1 μeV = 242 MHz = 4 ns

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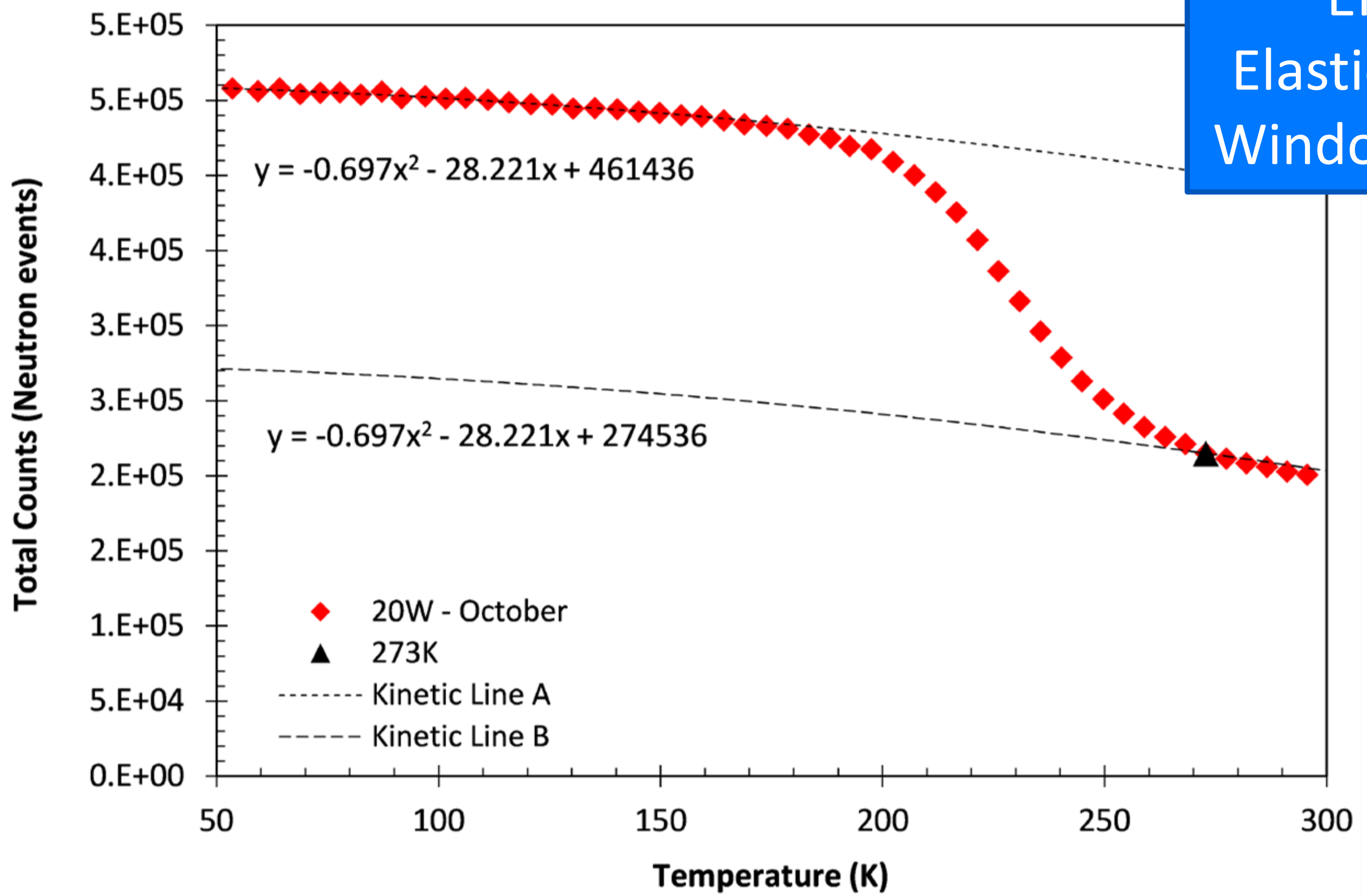
Science on EMU: Case study 1

- Unfrozen Water In Na-montmorillonite



EFW
Elastic Fixed
Window Scan

Gates et al. P6167 (Manuscript in preparation)

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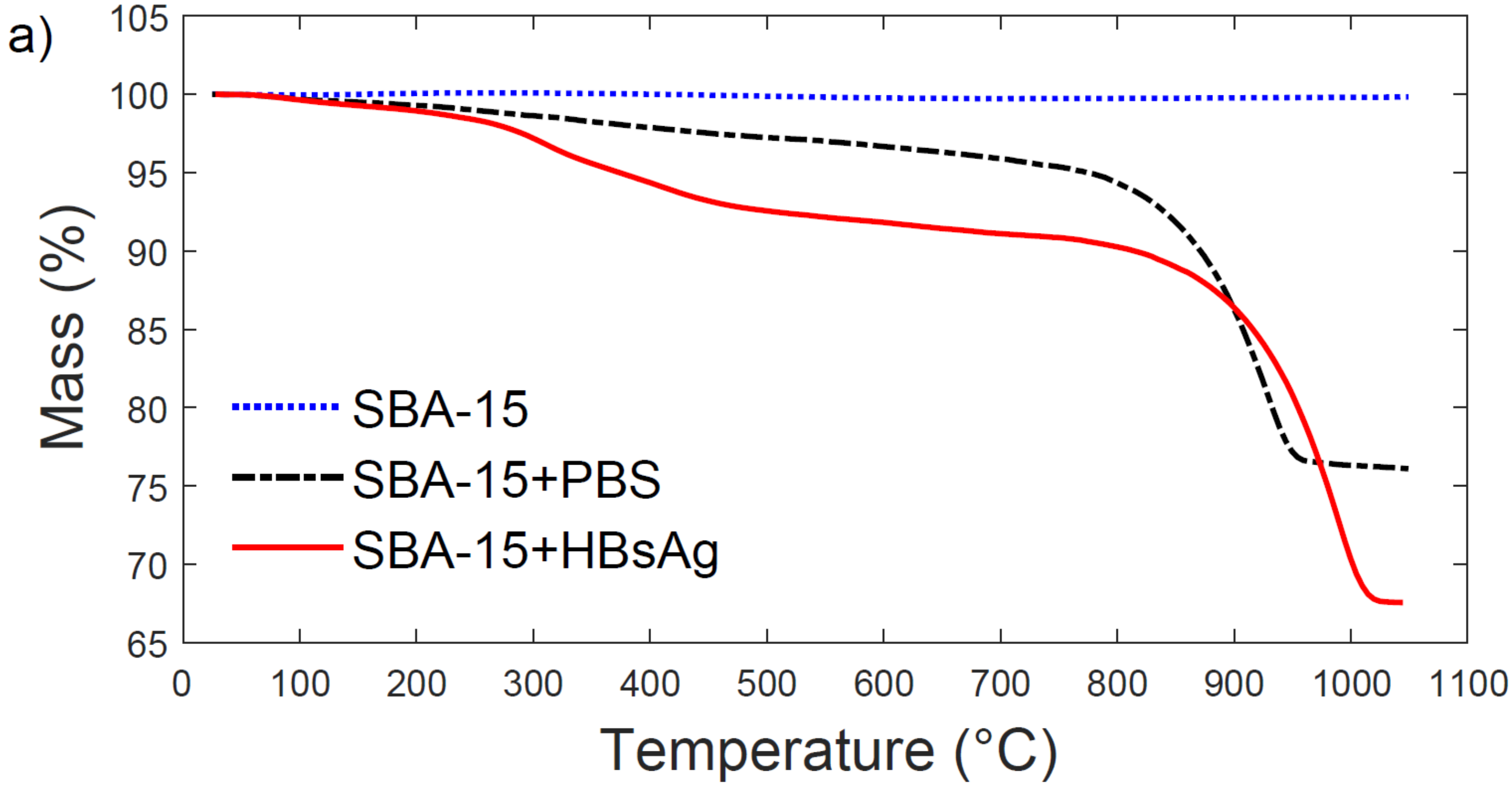
Science on EMU: Case study 2

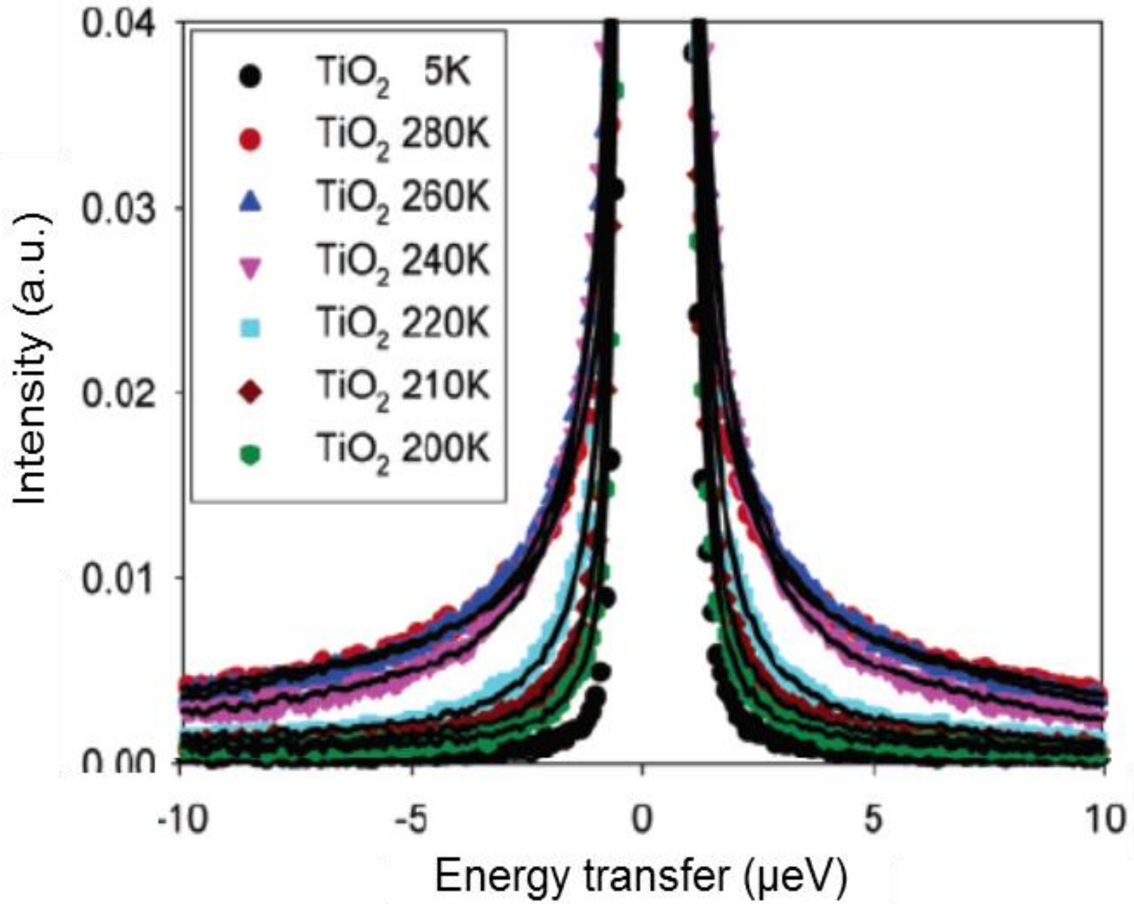
- Dynamics of encapsulated Hepatitis B surface Antigen



Rasmussen et al. EPJ Special Topics (Accepted, 2018)

Science on EMU: Case study 3

- QENS study of propane diffusion in gas hydrates
- Hydration water dynamics on rutile nanoparticles



E. Mamontov et al.
J. Phys. Chem. C. ***111*** *(2007)*

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EMU Capabilities

Science

- **Dynamics** of soft condensed matter such as polymers, proteins, biological membranes and gels
- Local and long range diffusion of liquids, solutions and confined systems
- Properties of quantum liquids, Fermi and non-Fermi systems

Sample Environment

- Gas-loading
- Standard cryostat temperatures
- Dilution temperatures

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Acknowledgments

PELICAN

Dehong Yu

Richard Mole

EMU

Nicolas de Souza

Alice Klapproth

RMIT UNIVERSITY

Thank you for your attention!

gail.iles@rmit.edu.au

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