(2018 -2028) Diffraction : Evolution, Révolution !

(Neutrons)

F. Porcher - LLB

LOOKING BACK : 50 YEARS OF NEUTRON SOURCES FOR RESEARCH

Cost/Technics :
National or International

NEUTRON SOURCES FOR RESEARCH: INTERNATIONAL LANDSCAPE (2018)

No small (local) source available for neutrons users

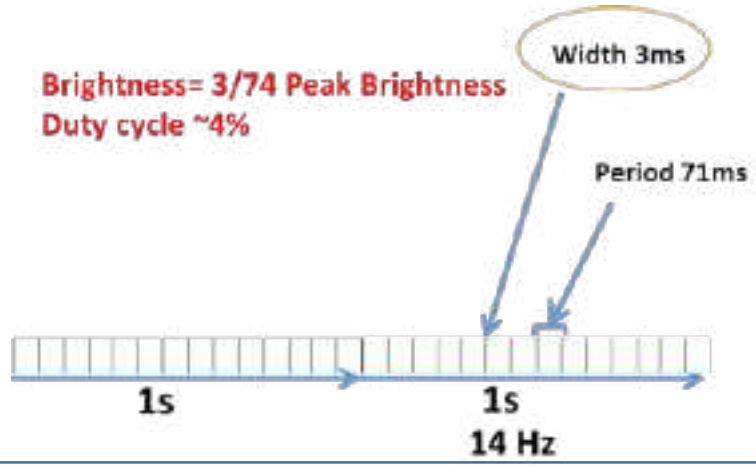
~30 sources for fundamental research (neutron scattering)

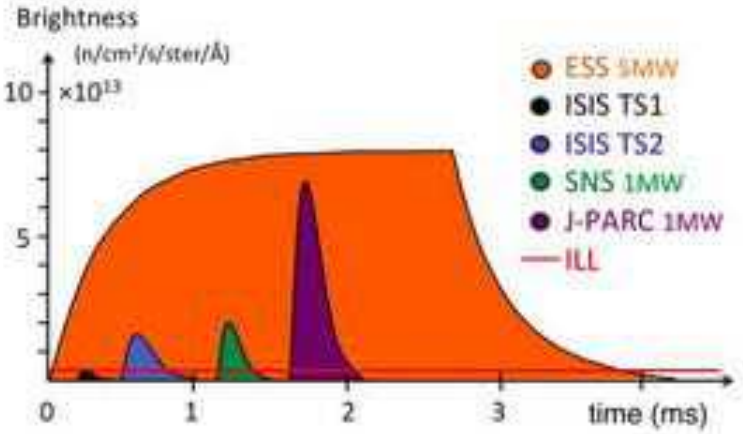
~15 high/medium neutron flux ➔ diffraction → **11 Reactors**
→ **4 Spallation sources**

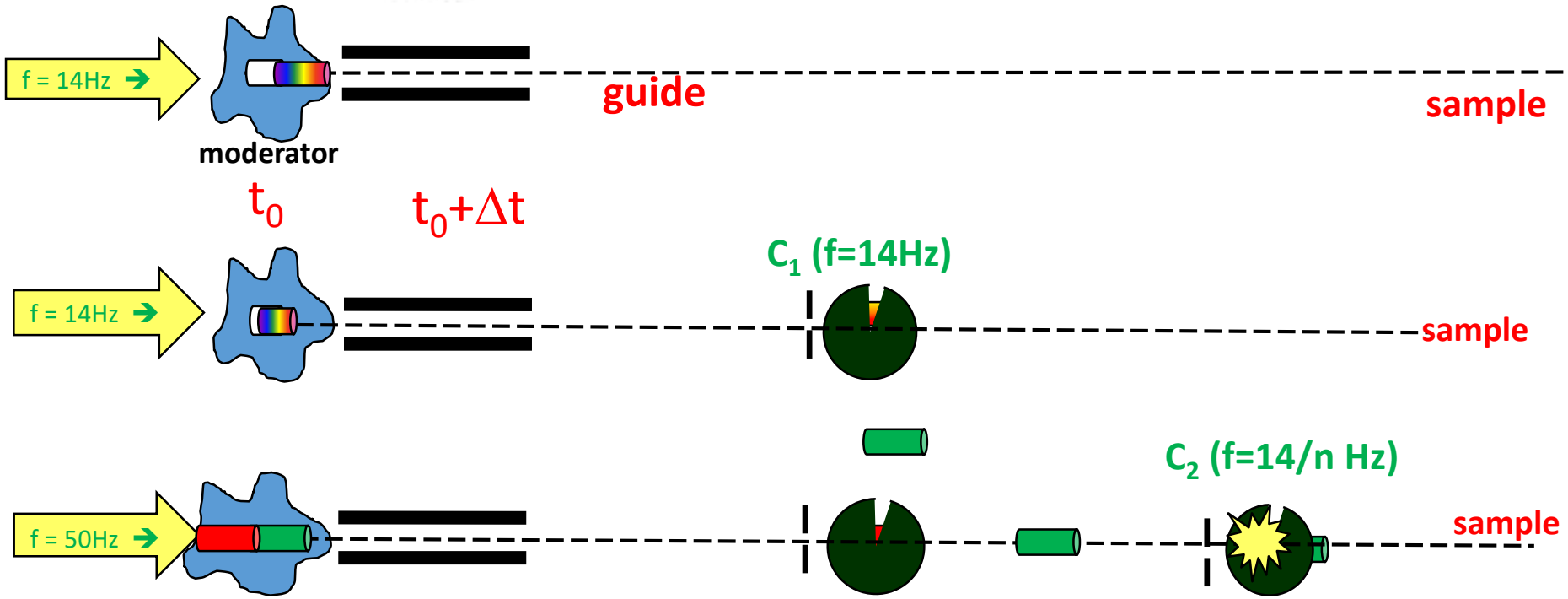
SYNCHROTRONS : INTERNATIONAL LANDSCAPE (2018)

Thousands of lab. machines for everyday Science & characterization

STEADY/PULSED NEUTRON SOURCE

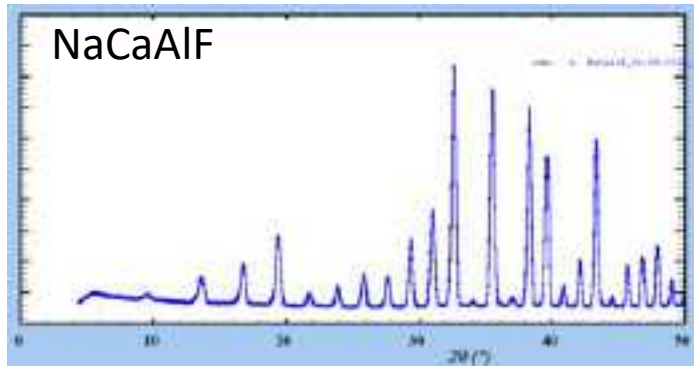






REACTOR vs SPALLATION SOURCE ~ CONSTANT WAVELENGTH vs TOF DIFFRACTION

Steady source



scattering angle (°)

$2d_{hkl} \sin\theta = \lambda$

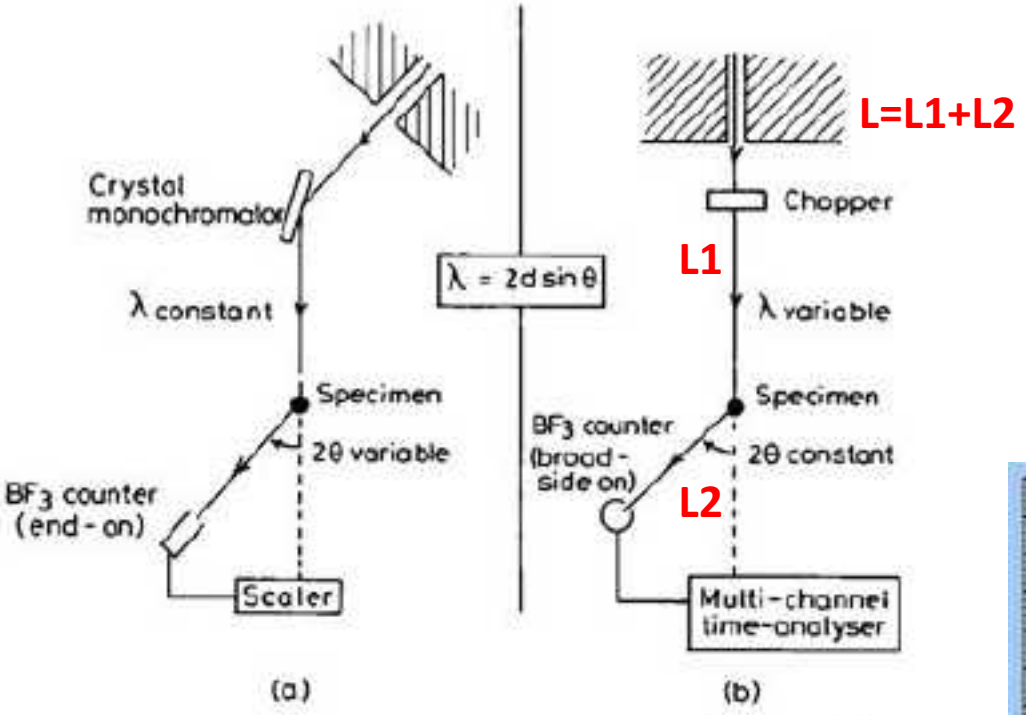
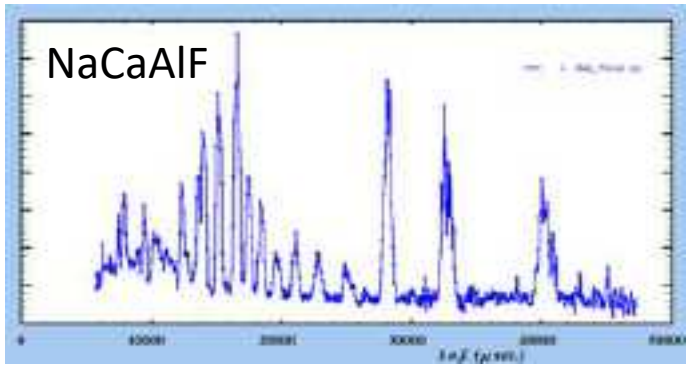


Fig. 1. The principles of (a) the reactor method (b) the time-of-flight method for crystallographic studies (after Buras, Leciejewicz, 1964).

Resolution : $\delta\lambda \sim \delta d_{monok}, \delta\theta_B$

Pulsed source



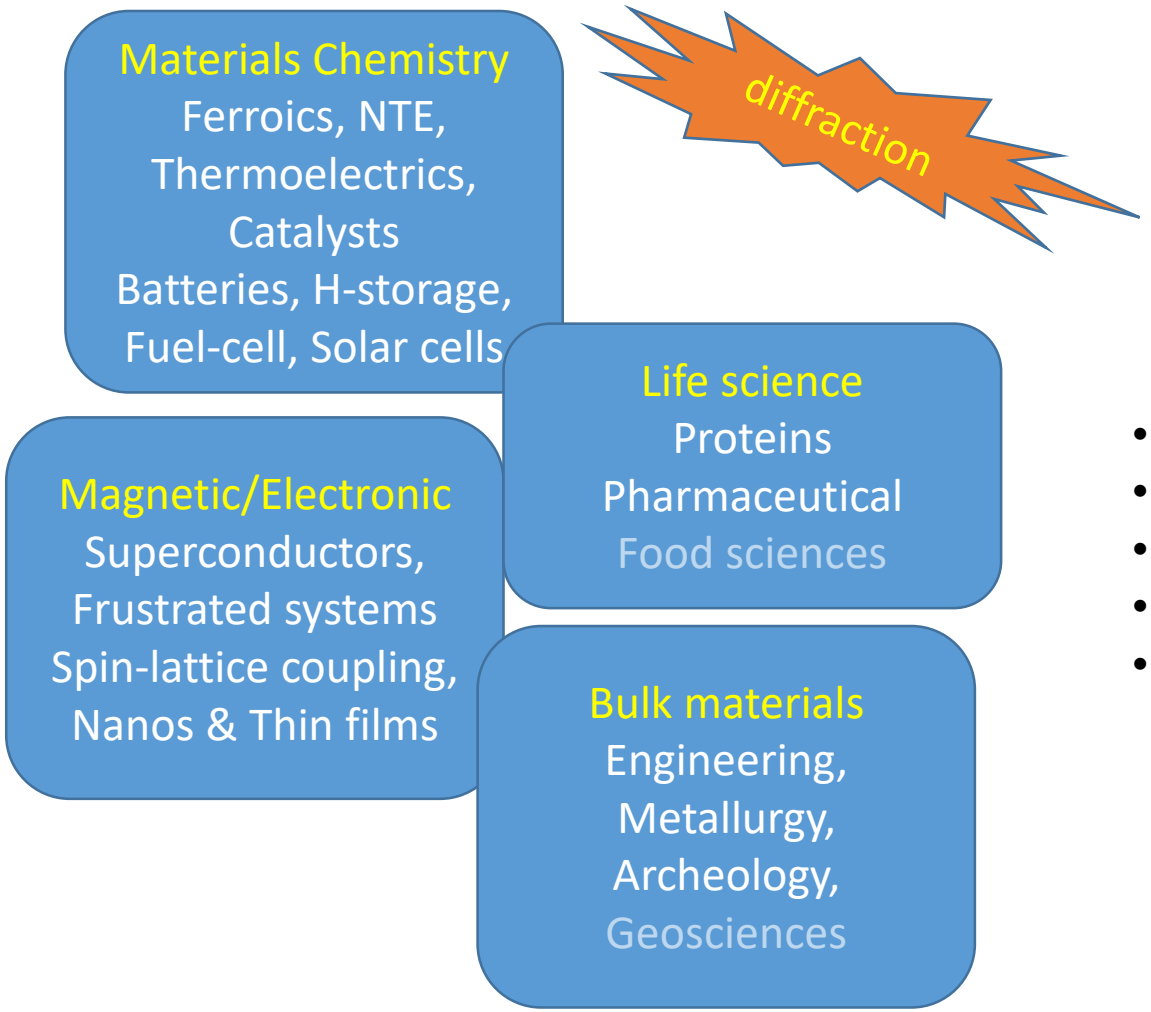
delay (µs)

Resolution : $\delta\lambda \sim \delta t_0, \delta t, L$

LOOKING FORWARD : WHY NEUTRON SCATTERING IN 2018 -

THEMA

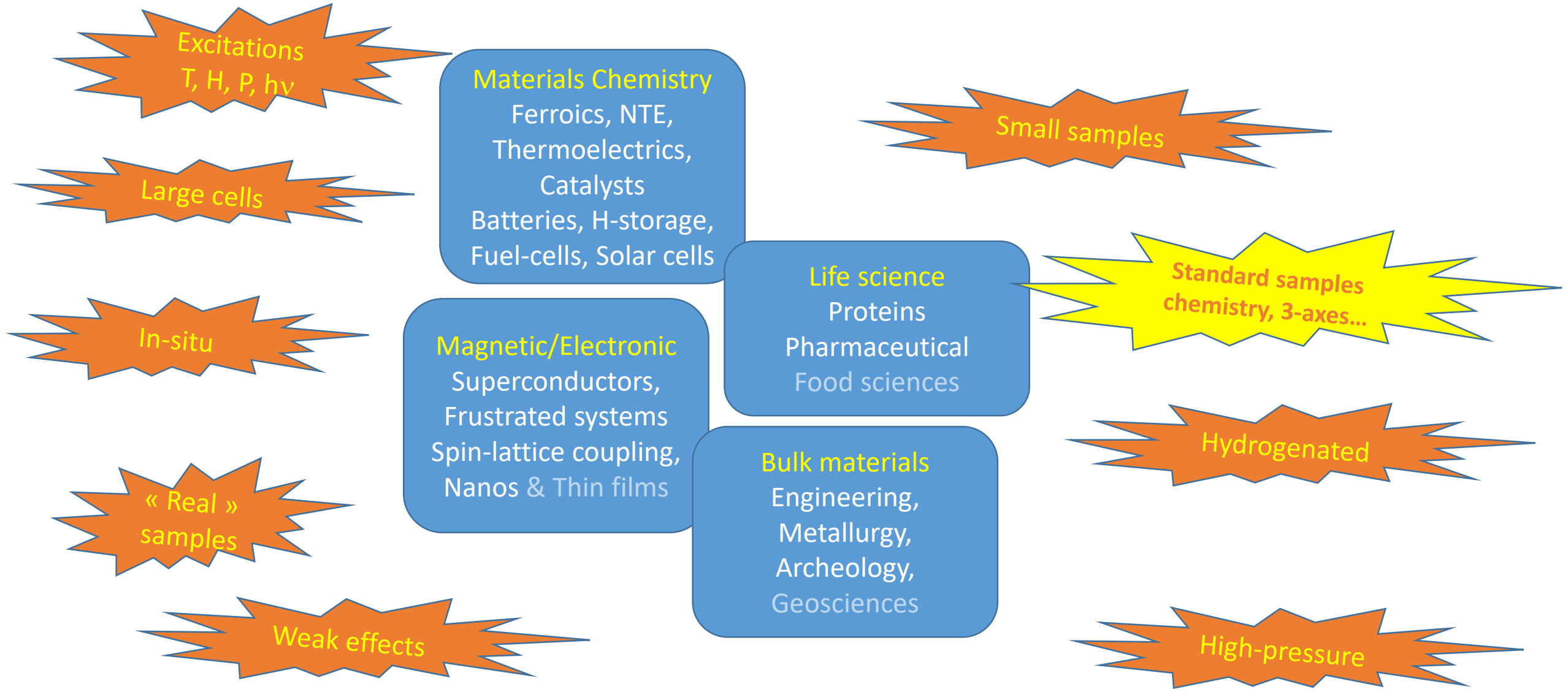
- **Broad Science**
- **Science for Society**
- **Industry**
- **Hot Science**



SPECIFICITY / X-Rays

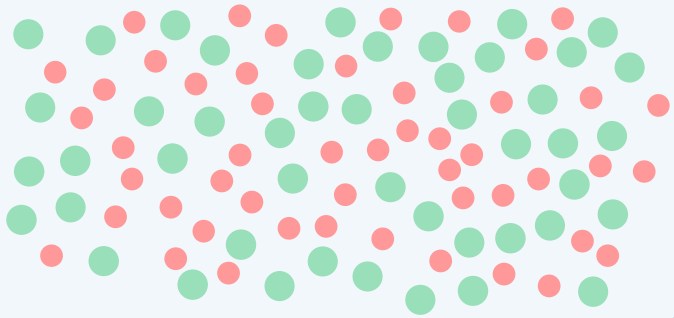
- **Probe structure & Motion**
- **Nuclear contrast (H, 3d elements)**
- **Magnetism**
- **High penetration**
- **Isotope labelling**

LOOKING FORWARD : WHY KIND OF DIFFRACTION EXPERIMENTS IN 2018 -



« DIFFRACTION » METHODS IN CONDENSED MATTER

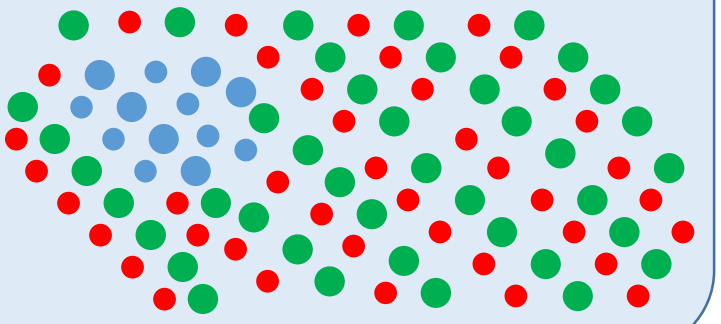
DISORDERED



Liquids, Glasses
➔ VdW glasses…, alloys)

Pair Distribution Function
(PDF)

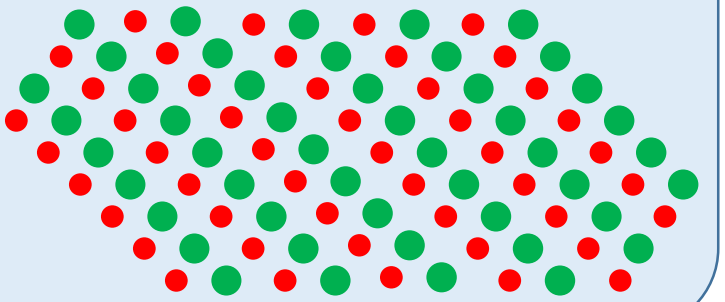
SHORT RANGE ORDER



Nanoparticles, Defects
➔ Catalysts, Batteries,
In-situ, Magnetic domains

PDF, Diffuse scattering,
Diffraction (profile analysis)
SANS

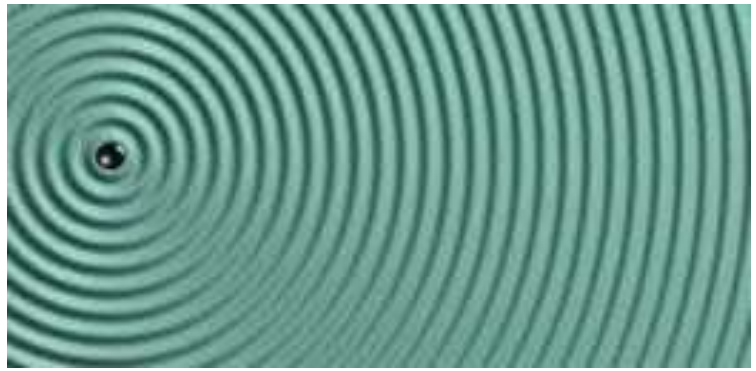
CRYSTALLINE

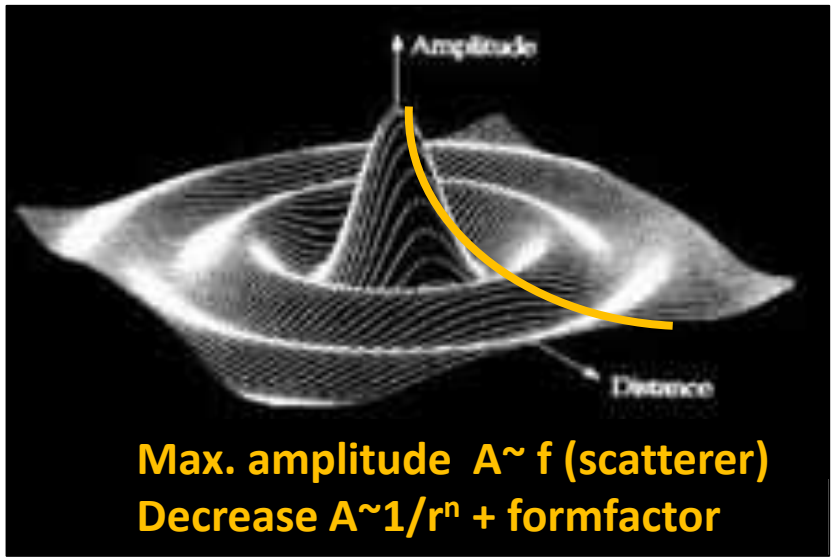


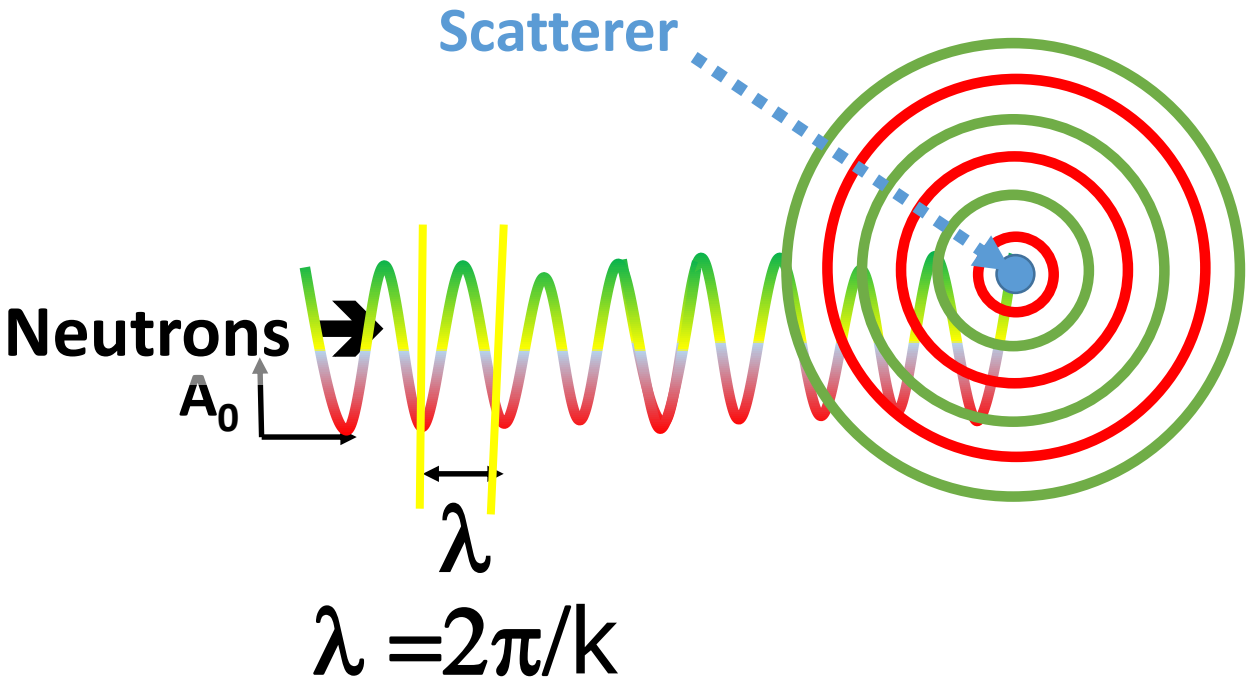
« Normal » crystalline materials
Intermetallics, Oxides, sulfides…
Pharmaceuticals, Proteins

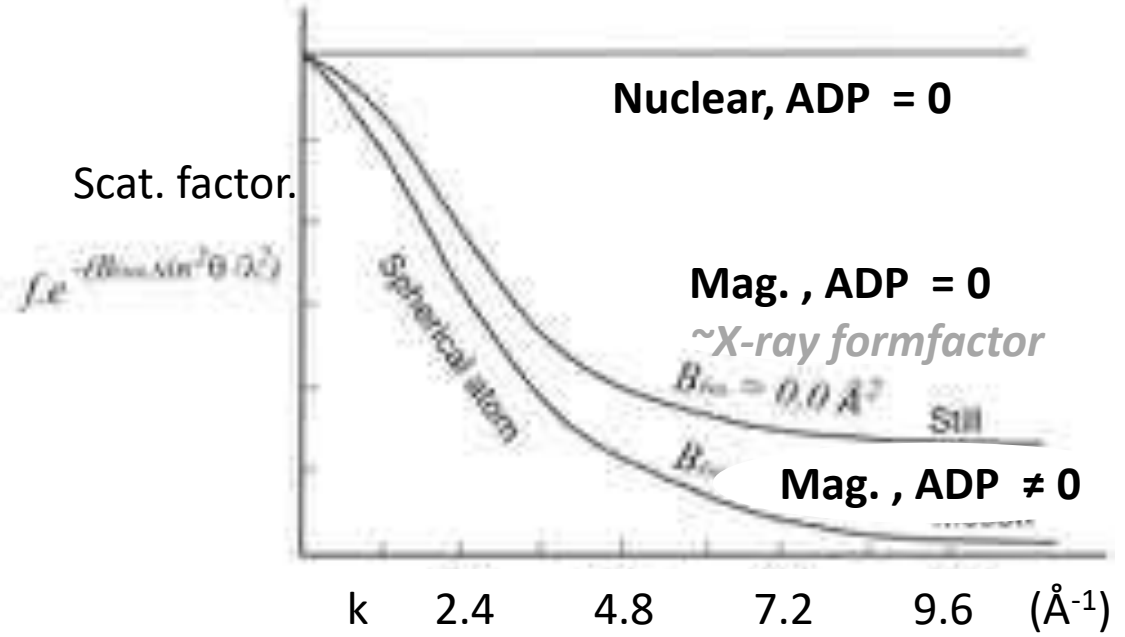
Diffraction
(SXD, Rietveld)

Prerequisites on data ? Back to the basics…

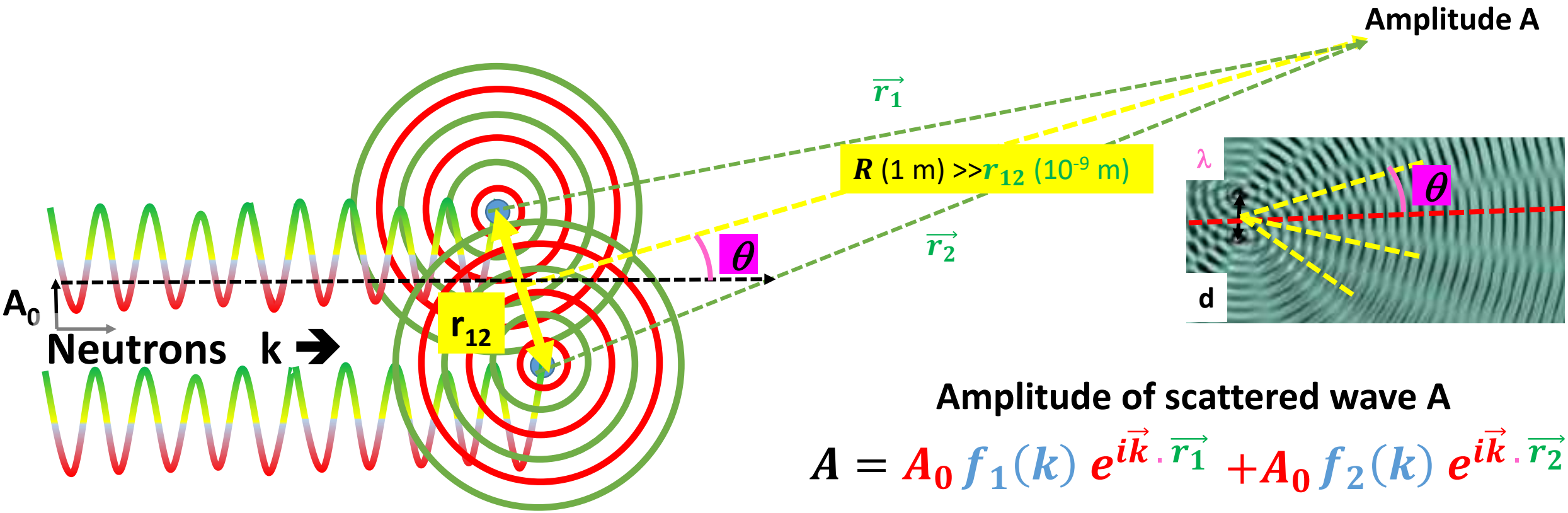








Back to the basics…



Amplitude of scattered wave A

$$A = A_0 f_1(k)\, e^{i\vec{k}\cdot\vec{r_1}} + A_0 f_2(k)\, e^{i\vec{k}\cdot\vec{r_2}}$$

$$A = A_0\, e^{i\vec{k}\cdot\vec{R}} \left(f_1(k) + f_2(k)\, e^{i\vec{k}\cdot\vec{r_{12}}} \right)$$

- A: Amplitude (~√signal on detector)
- A_0: Incident neutron flux
- $e^{i\vec{k}\cdot\vec{R}}$: Scattering angle θ (pos. detector, λ)
- $f_1(k) + f_2(k)\, e^{i\vec{k}\cdot\vec{r_{12}}}$: Structure Factor (atoms/moments pos.)

Path difference: $e^{i\vec{k}\cdot\vec{r_{12}}}$

Disordered materials : PDF

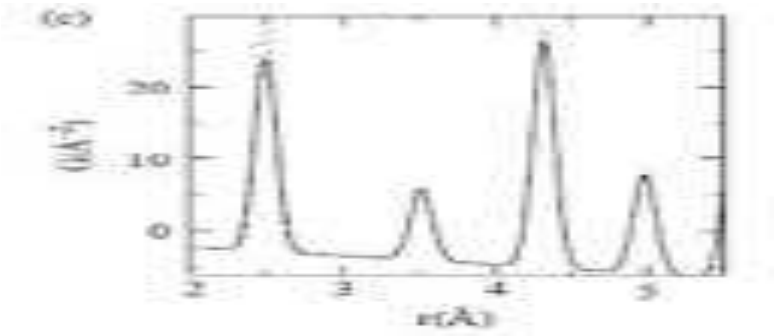
RECIPROCAL SPACE - SCATTERING

NORMAL SPACE

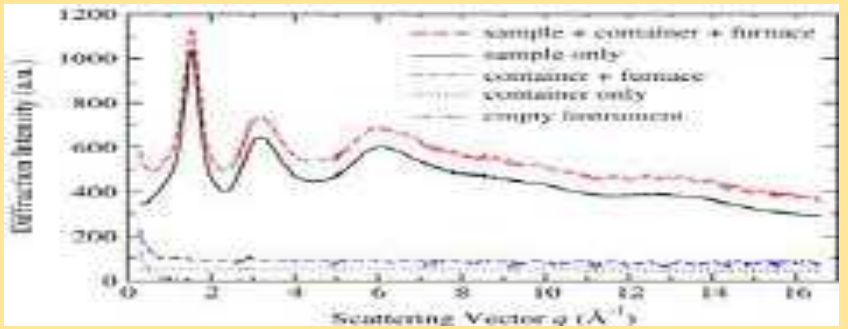
$I(\vec{k})$

r

k



Scattered Intensity I(k)



Isotropic

$$S(k) = \frac{I(k) - \langle f^2 \rangle - \langle f \rangle^2}{\langle f \rangle^2}$$

Weighting by k !

$$F(k) = k(S(k) - 1)$$

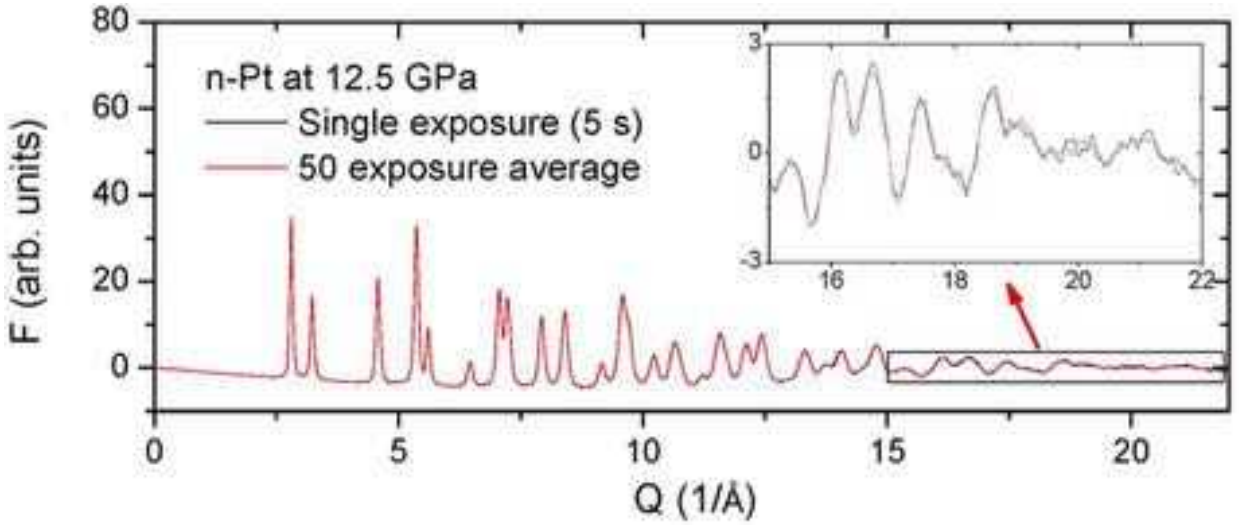
Pair distribution function G(r)

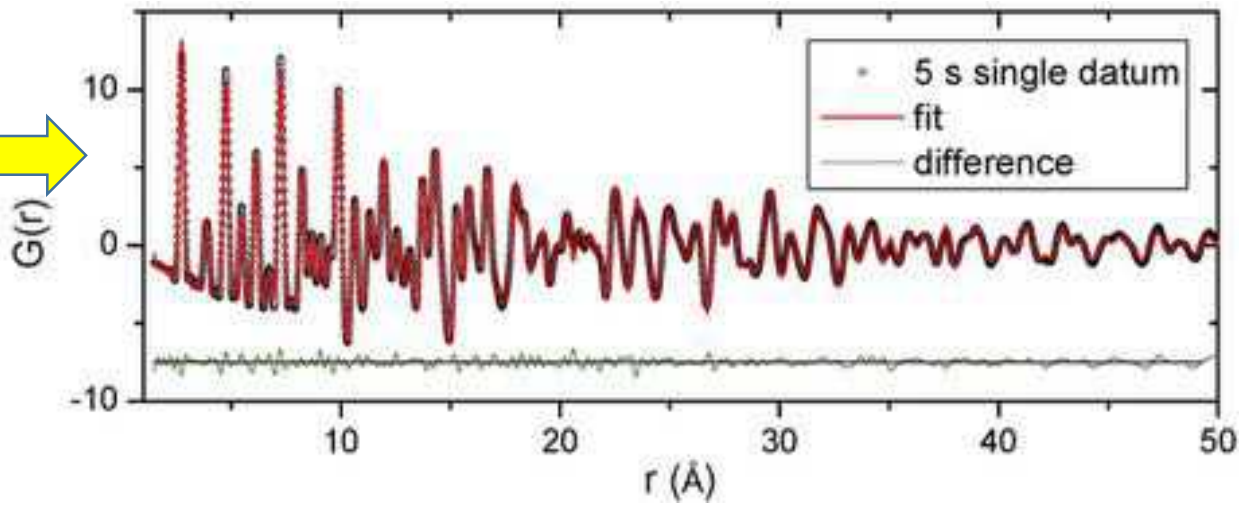
$$G(r) = \int_{k\,min}^{k\,max} F(k)\, sin(kr) dk$$ ← mesurement

$$G(r) = 4\pi r\,(\rho(r) - \rho_0)$$

$$\rho(r) = \frac{1}{4\pi r^2}\frac{1}{N}\sum_n \sum_{m \neq n} \frac{b_m b_n}{\langle b^2 \rangle}\,\delta(r - r_{mn})$$ ← structure

Experimental requirements for PDF…

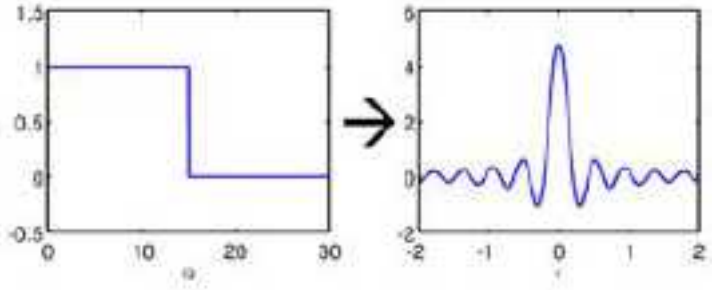


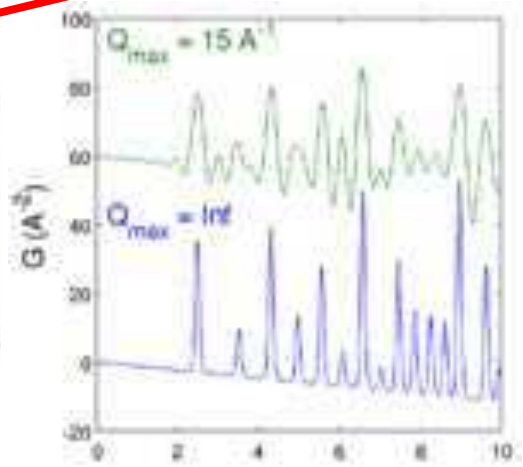


Pair distribution function G(r)

$$G(r) = \int_{k\,min}^{k\,max} k\,(S(k) - 1)\,sin(kr)\,dk$$

High Resolution k_{max}





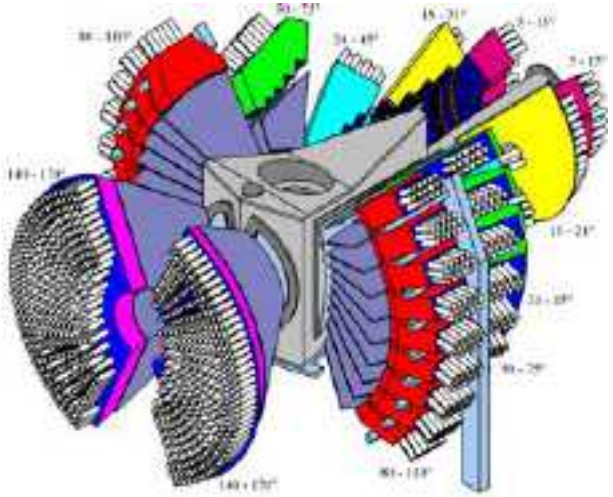
Intensity I(k) / S(k) / F(k) at high resolution k_{max}

Resolution Δk «fair» at k>>

TOF vs Monok

+ Flat/controlled background

Neutron diffractometers for PDF analysis



GEM@ISIS - k_{max}=40Å^{-1}

In-situ

Catalysts

Batteries

« Real » samples

Nanomag.

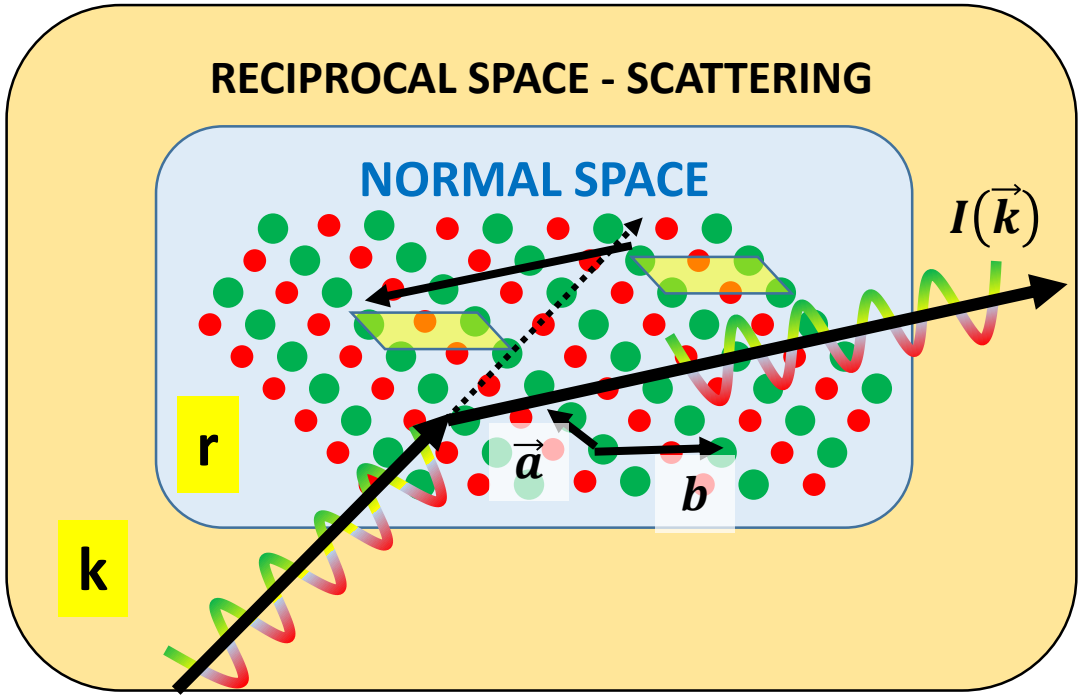
D4@ILL - k_{max}=33Å^{-1}

NOMAD@SNS - k_{max}=100Å^{-1}

7C2@LLB - k_{max}=20Å^{-1}

accessible to French community

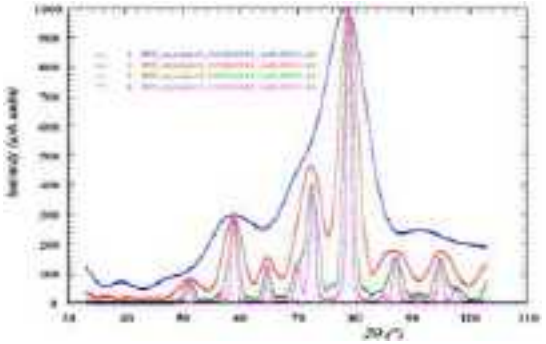
Crystalline materials : diffraction



Scattered amplitude ➔ diffraction

$$A(\vec{k}) \sim \sum_{N_a}\sum_{N_b}\sum_{N_c}\sum_{j\,(maille)} f_j\, e^{i\vec{k}.(\vec{r_j}+N_a\vec{a}+N_b\vec{b}+N_c\vec{c})}$$

$$I(\vec{k}) \sim \left(\sin\frac{(N_a\vec{k}.\vec{a})}{N_a\vec{k}.\vec{a}}\ \frac{\sin(N_a\vec{k}.\vec{a})}{N_a\vec{k}.\vec{a}}\ \frac{\sin(N_a\vec{k}.\vec{a})}{N_a\vec{k}.\vec{a}}\right)^2 \sum_{j\,(maille)} f_m f_n\, e^{i\vec{k}.\overrightarrow{r_{mn}}}$$



Interference function

Nanoparticles, clusters, stress/strain

Structure factor

Structures nuclear / magnetic

Crystallographics studies on powders : the k paradox

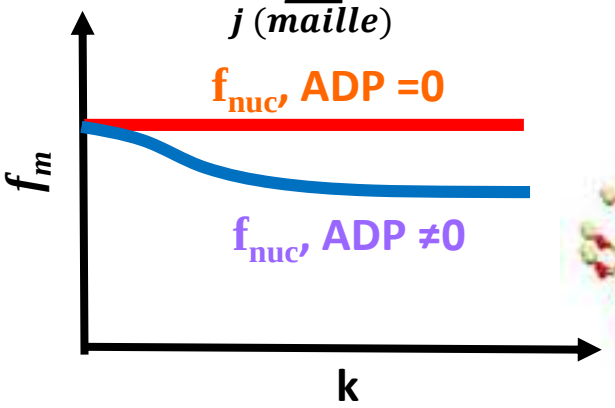
Catalysts

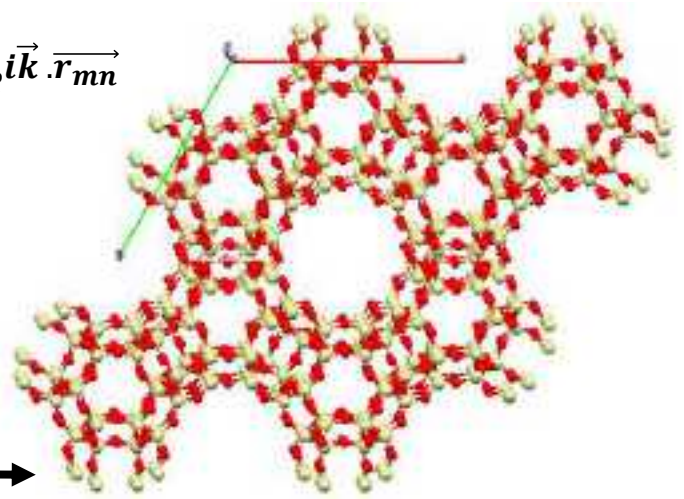
Pharmaceuticals

Nuclear structure of large cell compounds

- **Intense Bragg peaks at kmin**
- **Low scattering power at kmax**

$$I(\vec{k}) \sim \sum_{j\,(maille)} f_m f_n e^{i\vec{k}.\overrightarrow{r_{mn}}}$$



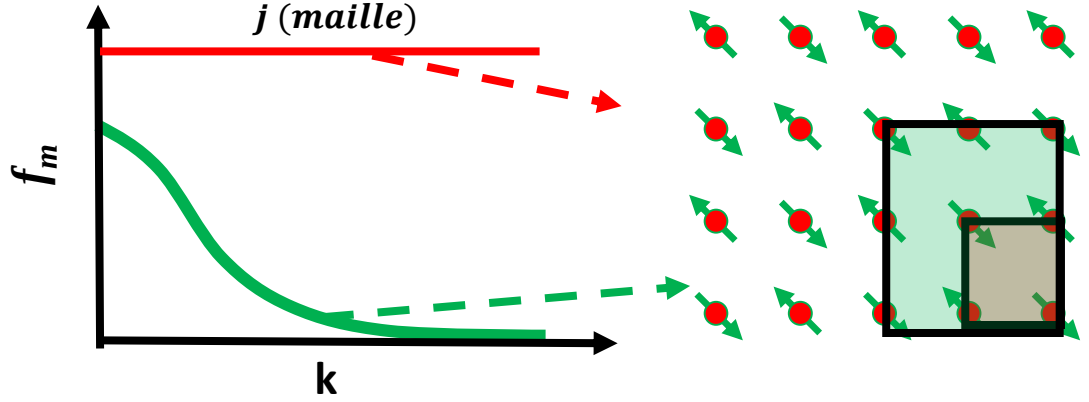


Magnetism

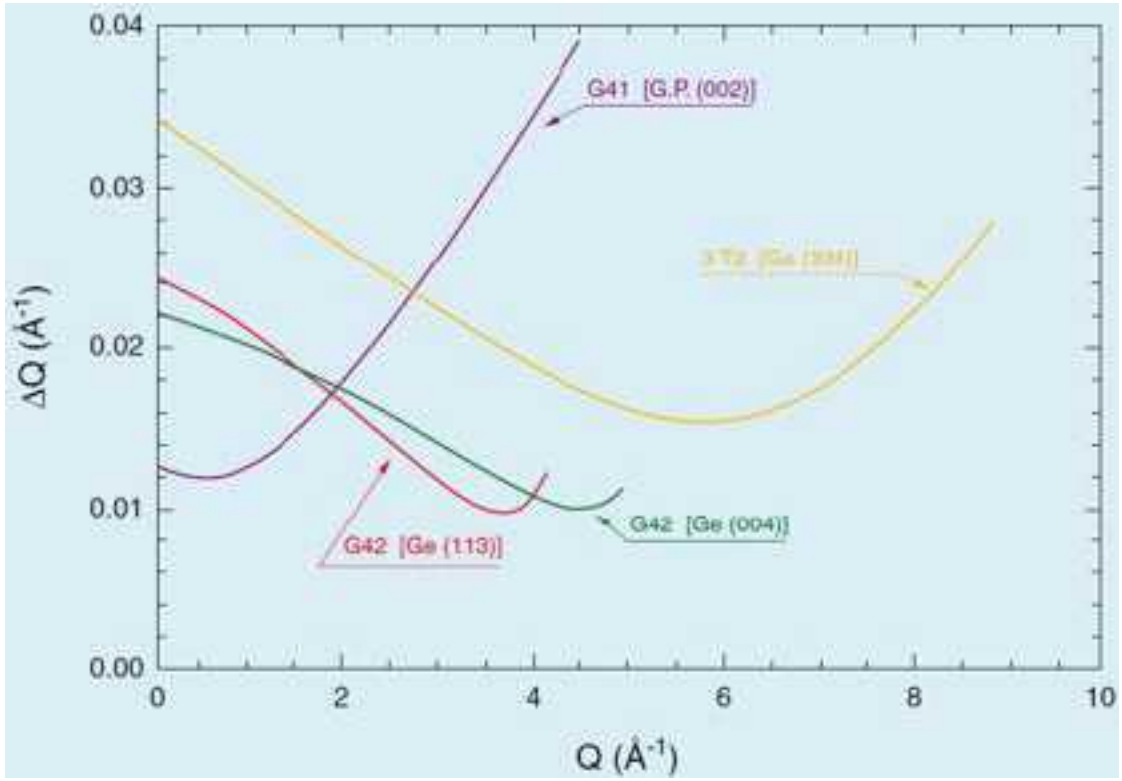
Magnetic structure

- **Magnetic peaks, diffuse scattering at kmin**
- **Data at kmax needed for nuclear structure**

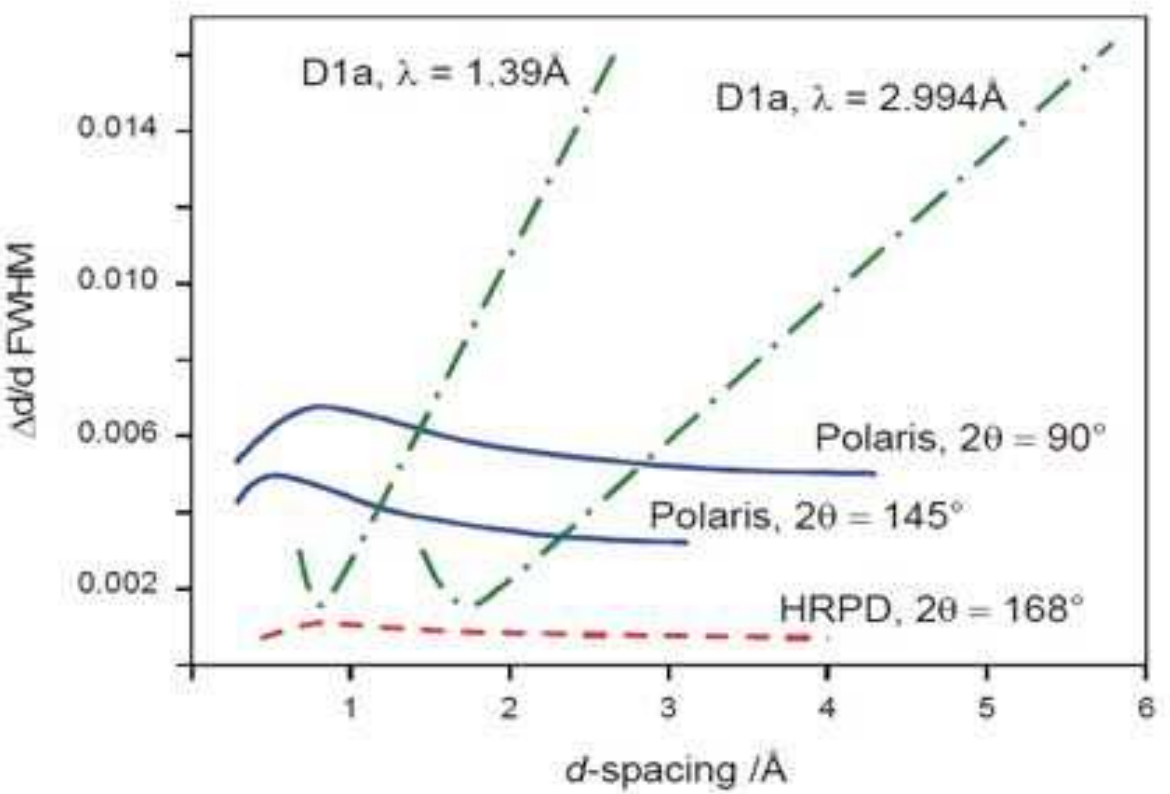
$$I(\vec{k}) \sim \sum_{j\,(maille)} f_m f_n e^{i\vec{k}.\overrightarrow{r_{mn}}}$$



Resolution(s) kmin, kmax, Δk in CW / TOF diffraction



CW instruments : 2θ, collimations, mosaicity, monok take-off



TOF instruments : 2θ bank, divergence, length, moderator

Resolution ⇔ Intensity

Peak profile TOF < CW

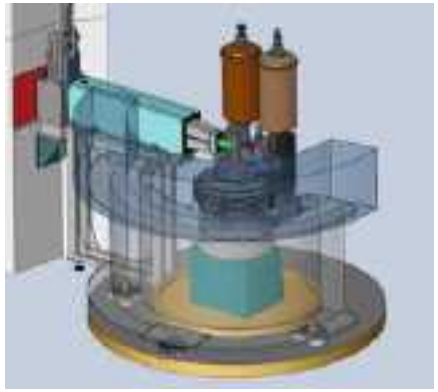
Combining diffractometers with CW – One shot experiment with TOF

High intensity

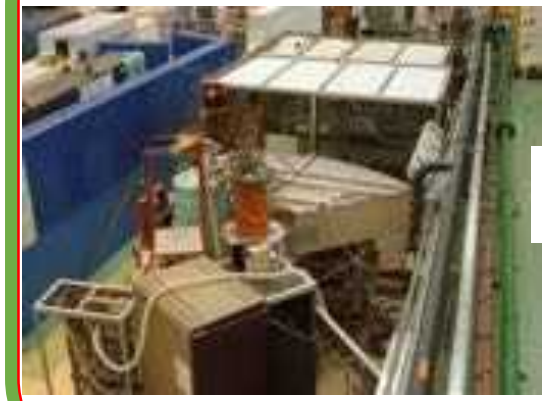
D1B@ILL

&

High resolution



D2B@ILL

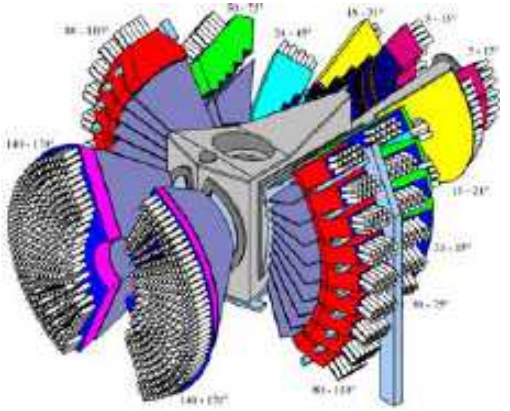


G41@LLB

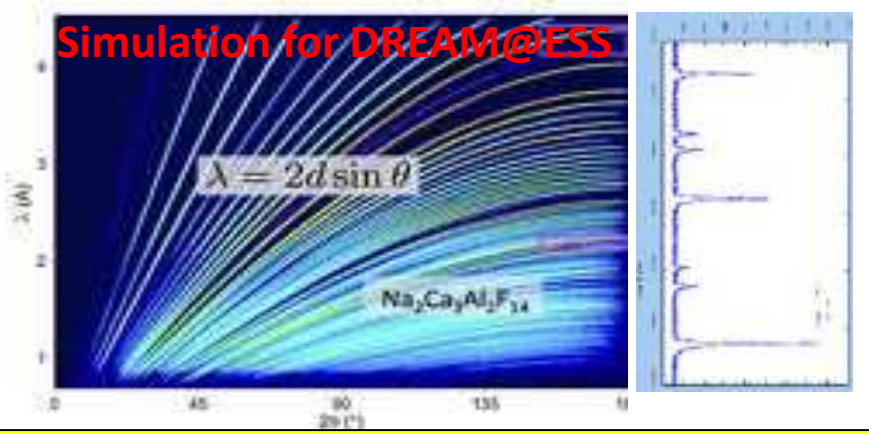
&

3T2@LLB

accessible to French community



2θ~180° : Backscattering
2θ~90° : Min. Background
2θ~0° : SANS, d_{max}



- Extensive ^{3}He detector banks (cost !!!)
- Background
- Normalization / data reduction
- Data analysis in progress

Single crystal diffraction

Excitations
T, H, P, hν

Weak effects

Superconductors

Magnetism

$2d_{hkl} \sin \theta = \lambda$

+ neutron polarisation

1

2

3

CW + Goniometer

Monochromatisation
+ motor rotations

Classical (X-ray type) data reduction

Laue (1912, X-ray)

Wide $\Delta\lambda$ band
Almost static (ω rotation)

No lambda/TOF control
➔ Tricky data reduction

Laue TOF (neutrons only !)

Wide $\Delta\lambda$ band
Almost static (ω rotation)

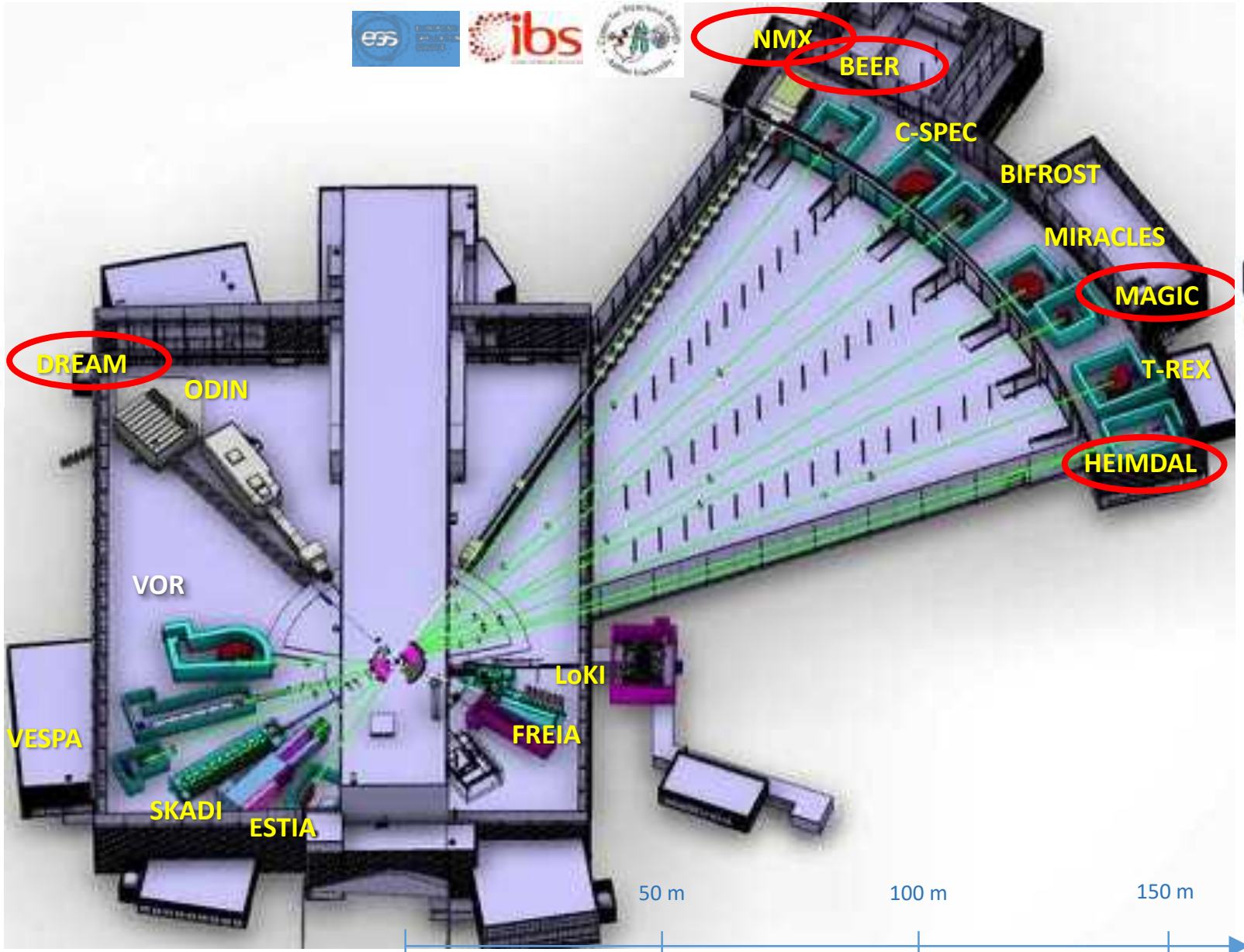
TOF data reduction in progress

FRENCH/EUROPEAN ACCESS TO DIFFRACTOMETERS : 2018-2028

Shutdown of Orphée (2019) / SOuP of ESS (2024-) / ILL ? / Sonate ?

LLB (french)		ILL (European) (french CRG)		ESS (European)		
G4.1	Powder, high flux, cold	D1B	Powder, high flux, cold (thermal)	DREAM	Non polarised	Powder >> Crystal
G4.4	Powder, high resolution, cold	D2B	Powder, high resolution, thermal			
3T2	Powder, high resolution, thermal	D20	Powder, high flux, thermal & cold	Heimdal	Non polarised	Powder
7C2	Amorphous, high flux, hot	D4	Amorphous, high flux, hot			
		D7	Diff. scattering, polarised, cold			
5C1	Crystal, polarised, hot	D3	Crystal, polarised, hot	MaGiC	Polarised	Crystal >> Powder
5C2	Crystal, non polarised, hot 4-circles	D9	Crystal, non polarised, hot 4 circles			
6T2	Crystal, w/o polarised, thermal	D10	Crystal, non polarised, cold 4 circles			
		D19	Crystal, non polarised, thermal			
		D23	Crystal, w/o polarised, thermal			>2019
		Cyclops	Crystal, Quasi-Laue			
		LADI	Crystal, Quasi-Laue, protein	NMX	Non polarised	Crystal (Protein)
6T1	Engineering, thermal	SALSA	Engineering, thermal	BEER	Engineering	

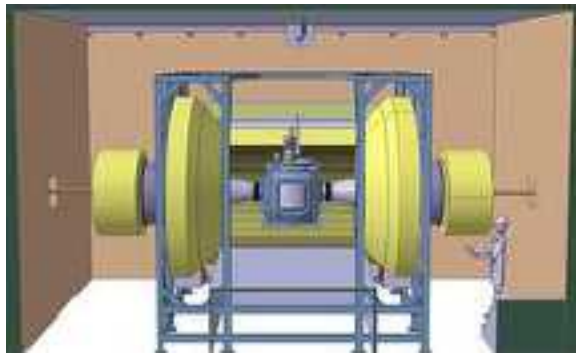
DIFFRACTOMETERS @ ESS



Diffractometer

FRENCH CONTRIBUTIONS TO DIFFRACTION AT ESS

DREAM

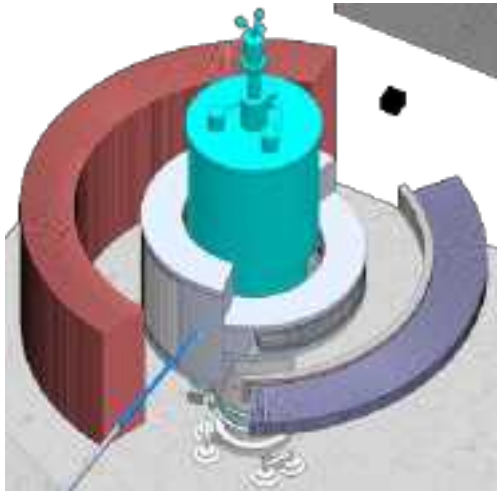


Powder diffraction

- Sample size down to 1mm^3
- Time resolution down to 1 ms (0.5cm^3)
- Q_{max} up to 25 Å^{-1}
- Q_{min} down to 0.01Å^{-1}
- Δd~0.0003 Å in BS (see ID31)

Single crystal diffraction (unpol.)

MaGiC



Polarised single crystal diffraction & Diffuse scattering

Sample size:

- Thin films: 1 cm2
- Bulk: < 1mm3 (down to 0.001 mm3)

Lattice parameters:

- Crystal: 5-30 Å
- Magnetic: 5-200 Å

NMX

Single crystal diffraction (unpol.)

- Small crystals (~200 µm)
- Fast data collection (days)
- Large unit cells (up to 300 Å)
- Robotisation

COMPACT ACCELERATOR-BASED NEUTRONS SOURCES

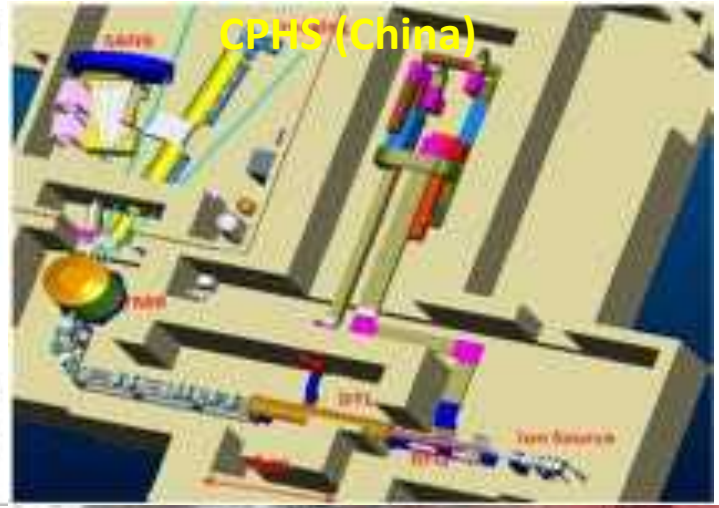
- CRGs / Access time on reactors (ILL, FRMII) ➔ CW diffractometers (maintain communauty)
- New pulsed neutron source ➔ TOF diffractometers (prepare communauty to ESS)

Spallation (ESS)

1 p (GeV) + 1 A(Hg, W)
➔ 15 n (MeV) + A' + γ

Stripping (CANS)

1 p (10 MeV) + 1 A(Be, Li)
➔ 0.05 n (MeV) + A' + γ



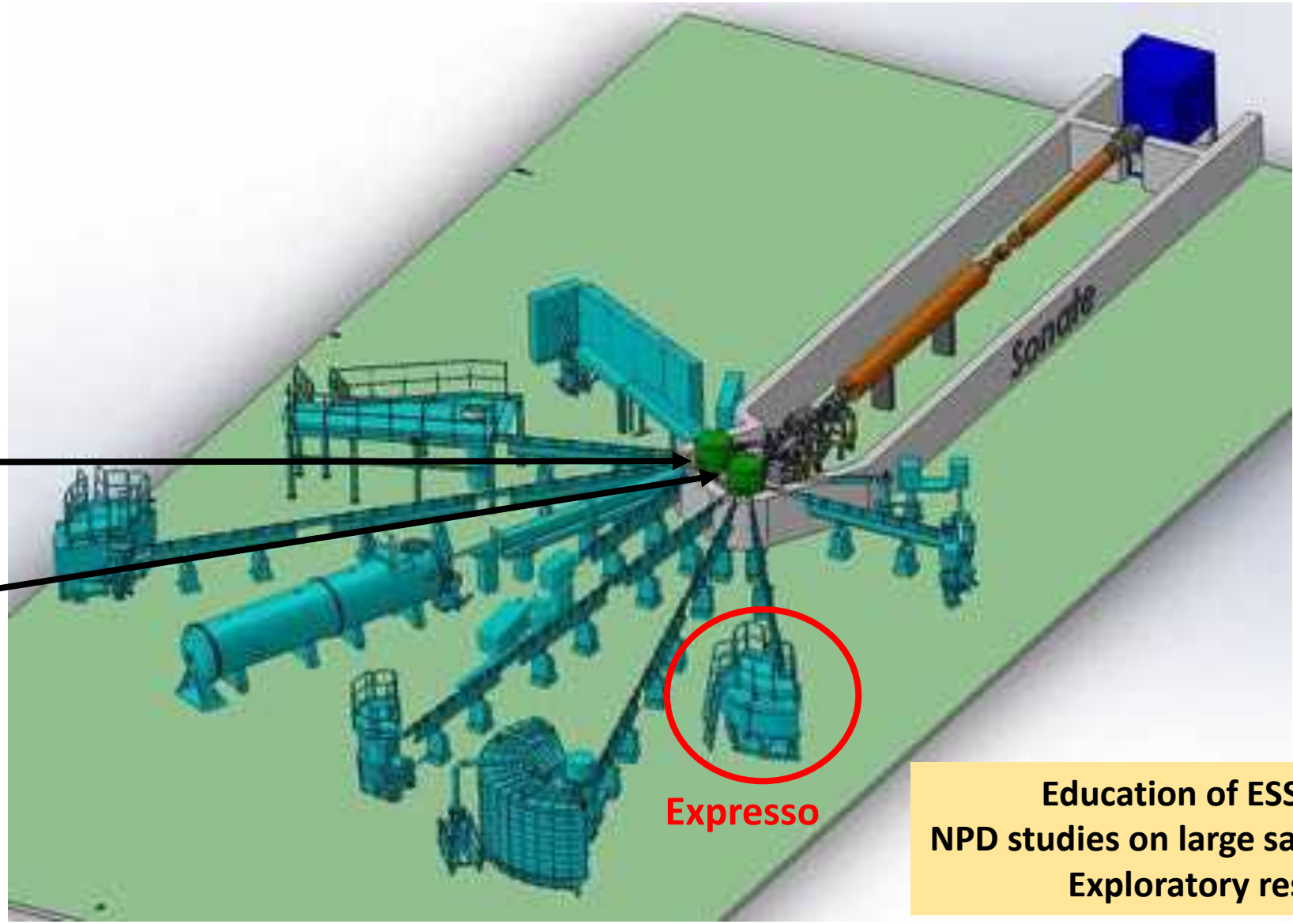
+ SONATE, German projects

LOOKING FORWARD : TOF POWDER DIFFRACTOMETER AT SONATE

Sonate
NEUTRONS FOR MATERIALS SCIENCE

Target 1 : 2ms pulse, 20 Hz
SANS, reflectometer, Imaging

Target 2 : 20µs pulse, 100 Hz
NPD, ToF spectroscopy



High brillance:
(beam tube closer to moderator)
Adjustable pulse length/repetition

Education of ESS users
NPD studies on large samples
Exploratory research

Merci

Thank you

Tack mycket