Péter Falus, Béla Faragó, Peter Fouquet, Dimitri Renzy
ILL, Grenoble

WASP the wide angle neutron spin echo instrument

ICANS, Dongguan, 1 November 2023

Important points

- ○ ILL is alive and well no technical reason to stop before 2045
- ○ NSE is not just Soft matter
- ○ WASP bridges TOF/BS range to small angle NSE range

HiRES 2023
Synergies in HIgh RESolution Spectroscopy

12-15
DEC
ILL, France

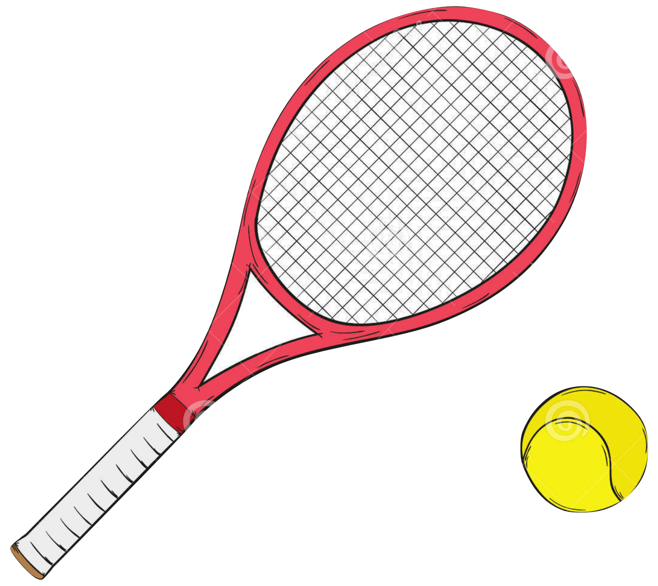
TOPIC

The workshop aims to bring together researchers in the field of neutron spin-echo, backscattering, and/or time-of flight (TOF) spectroscopy. The aim is to showcase the complementary of the techniques opening up new collaborations and attract new users. The scope of the workshop is not limited to neutron techniques, the participation of experts of complementary methods who wish to extend their research activity towards neutrons are welcome as well.

CONFIRMED SPEAKERS

Quentin Berrod, CEA Grenoble, FR
Juan Colmenero, UPV/EHU, ES
Peter Falus, ILL, FR
Bela Farago, ILL, FR
Bernhard Frick, ILL, FR
Rony Granek, BGU, IL
Andrew Jackson, ESS, SE
Margarita Kruteva, JCNS, DE
Fankang Li, ORNL, USA
Virginie Marry, Sorbonne Univ., FR
Goran Nilsen, ISIS, UK
Foivos Perakis, Stockholm Univ., SE
Romain Sibille, PSI, CH

Fundamentals of NSE



$$\hbar\omega = m\upsilon'^2/2 - m\upsilon^2/2$$

final velocity

initial velocity

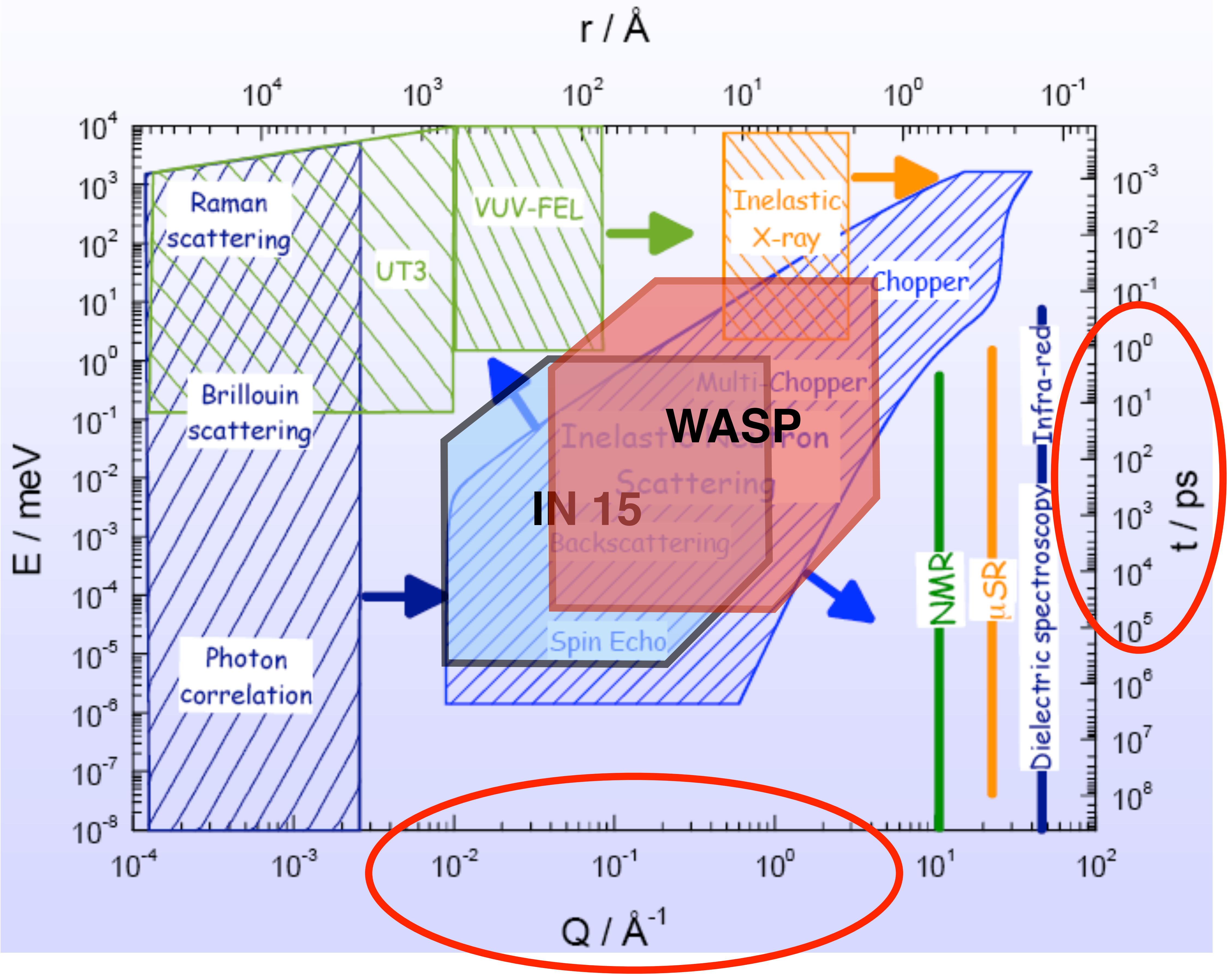
We want to measure the difference in velocity
The classical method is defining the final and initial velocity

Defining 2x== throwing away all other neutrons 2x
High resolution== very few neutrons remain ☹☹☹

Can we use all neutrons without defining/monochromatizing ?

Yes we can™ ! We will use the neutron spin. Spins rotate in magnetic field, we will encode the speed difference into number of rotations. (Feri Mezei 1972)

Why NSE important



- Unmatched Q-t range
- Works in time space not energy space
- Uses magnetic fields to encode speed difference
- Sees difference not sum of coherent and incoherent scattering
- For magnetism XYZ polarisation analysis built in

WASP is optimized for **atomic to molecular length scales 0.1-4 Å^{-1}, 0.2 ps-12 ns**

IN 15 is optimized for **molecular length scales 0.01-1 Å^{-1}, 5ps-1000 ns**

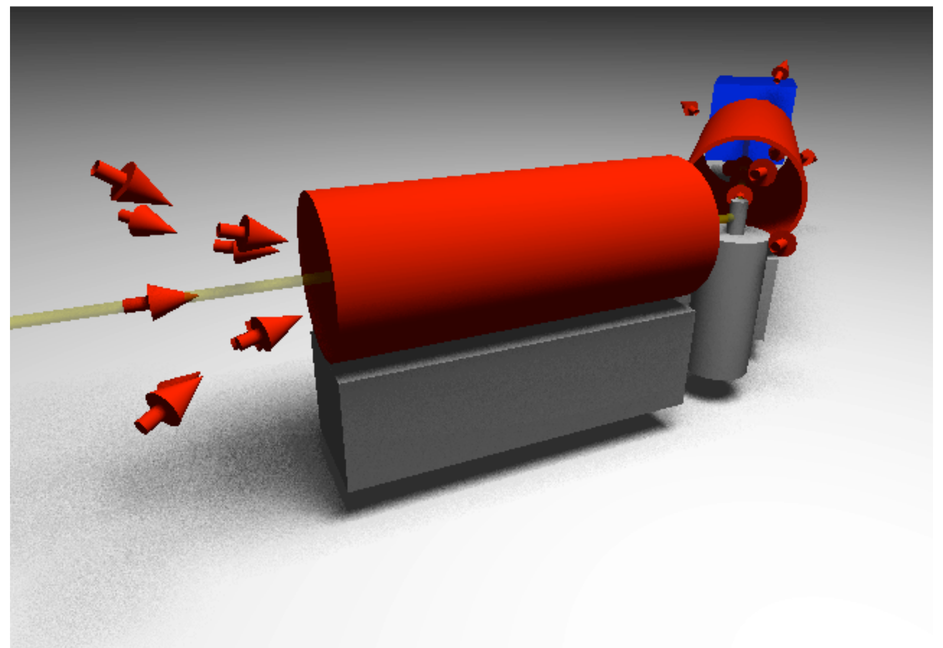
NSE Evolution

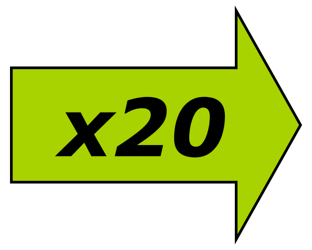
IN11A - high resolution
Mezei ’77, Farago ‘92

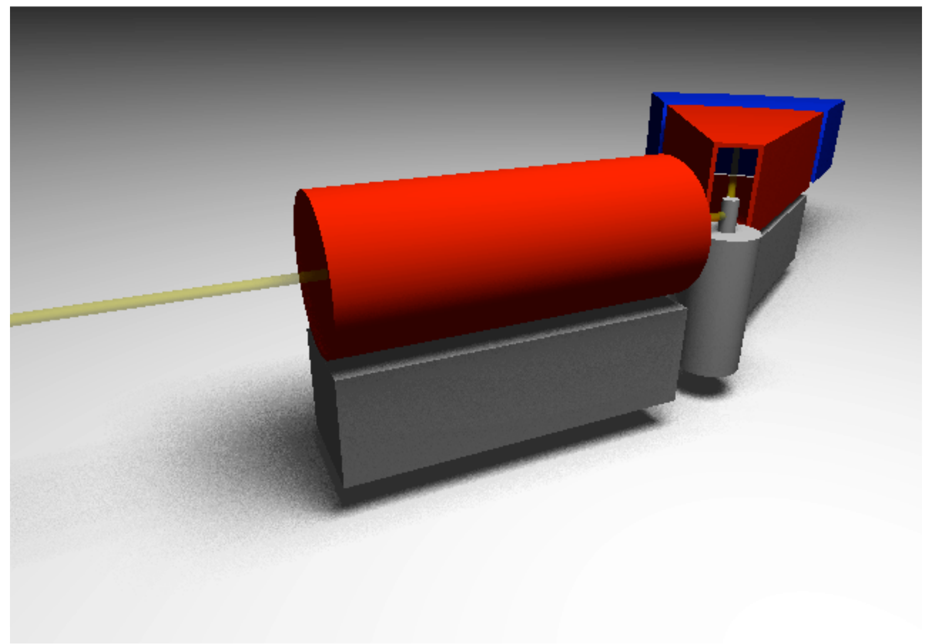
IN11C - 30º detector bank
Farago ‘97

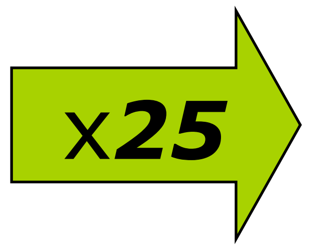
SPAN 30º detector bank
Mezei Pappas ‘99

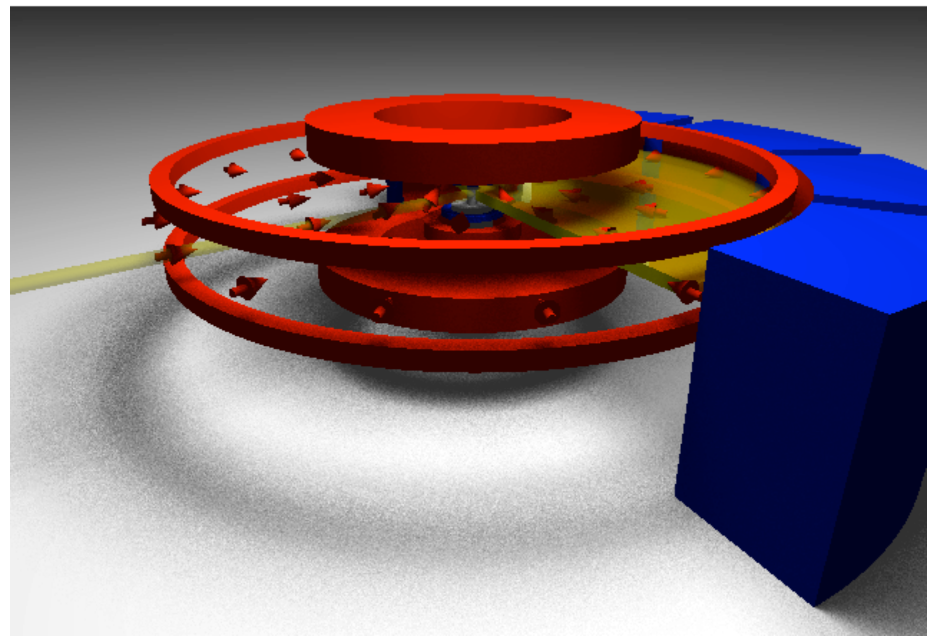
WASP 90º detectors ‘18











WASP will:

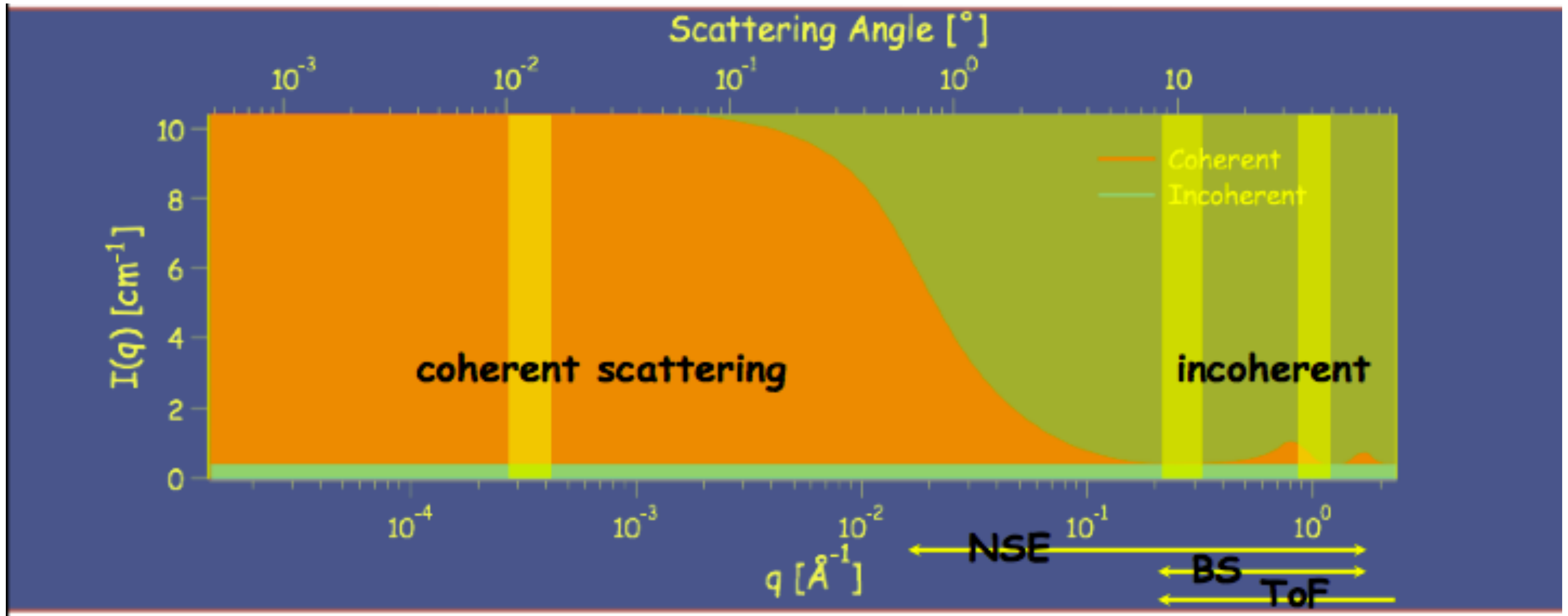
- Provide same **high resolution** as IN11A/ old IN15
- Increase the **sample flux x8**
- Increase the **detection solid angle x3** (90º compared to IN11C)

25x higher intensity **and** 6x higher field integral than IN11C

Reference: J. Neutron Res. 15, 39 (2007)

25x times intensity -paradigm shift

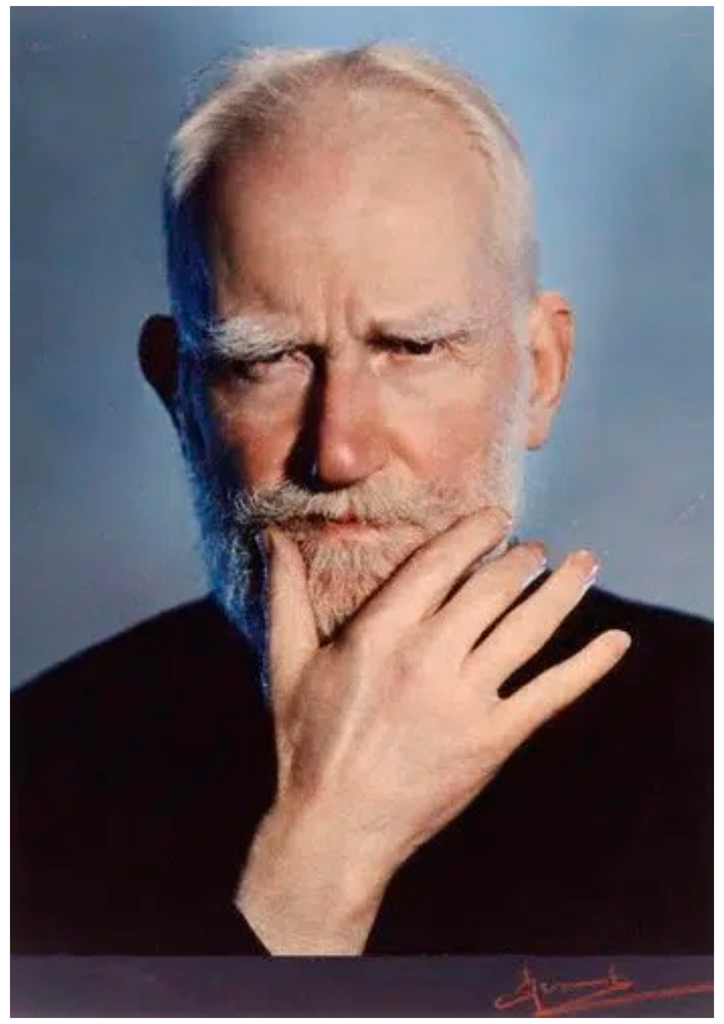
"Some people see things as they are and say why? I dream things that never were and say, why not?" G.B. Shaw



larger scale objects: slower dynamics

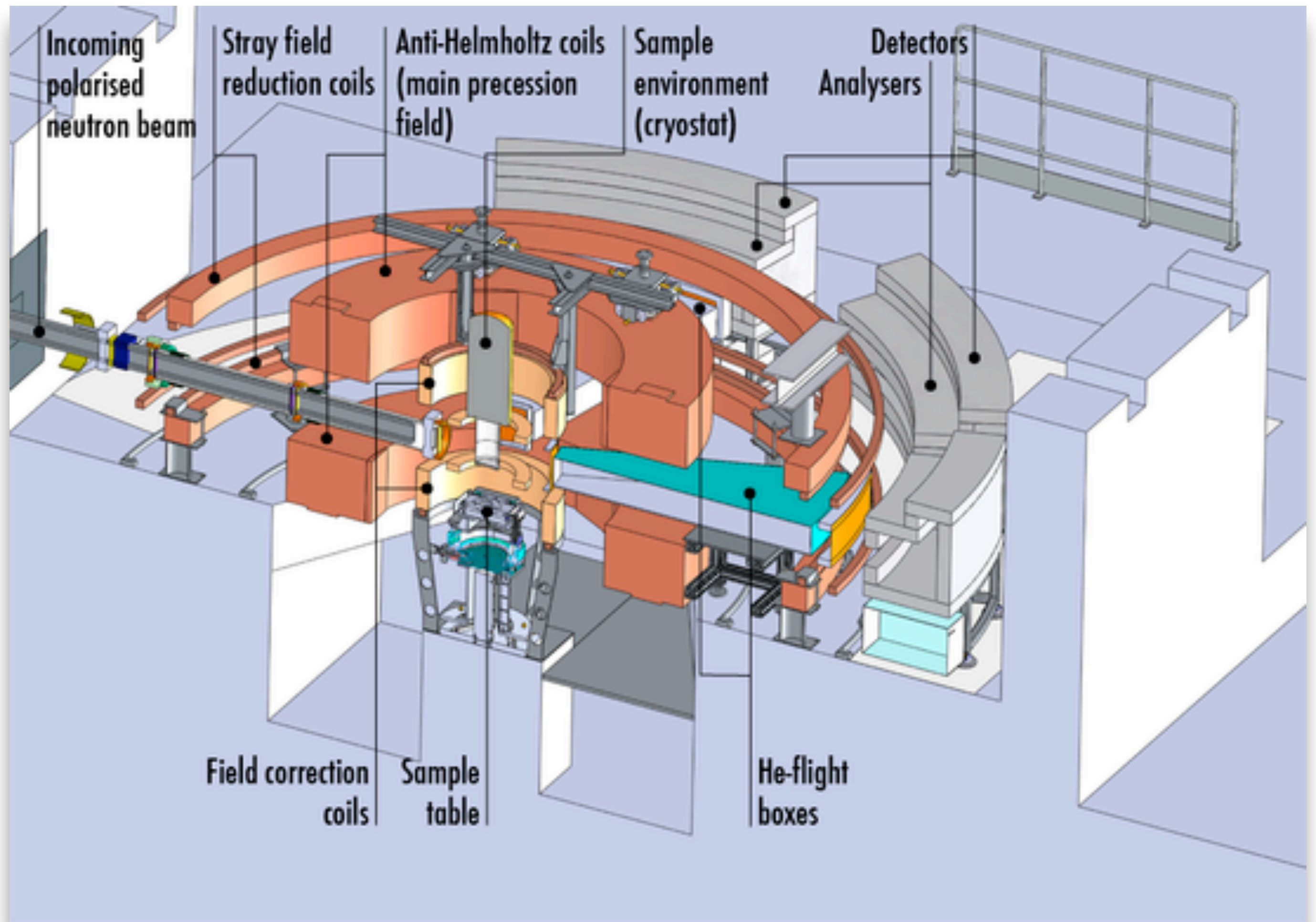
coherent dynamics at low Q and at Q corresponding to relevant length scales
incoherent dynamics at high Q

D. Richter, et al., *Adv. in polym. Sci.*, 174, 1 (2005).



Before WASP incoherent measurements took weeks, needed justification to invest beam time. Now there are results in hours.

WASP specs



First draft of proposal 2001

Proposal to instrument subcommittee 2005

Coil manufacturing 2015

Polarisation tests Apr 2018

First echo 4 October 2018

First users end of ~~2019~~ 2020

3+1 cycles alignment
5 cycles user operation

dynamic range
0.2 ps - 12 ns

0.05 - 3.5 Å^{-1}

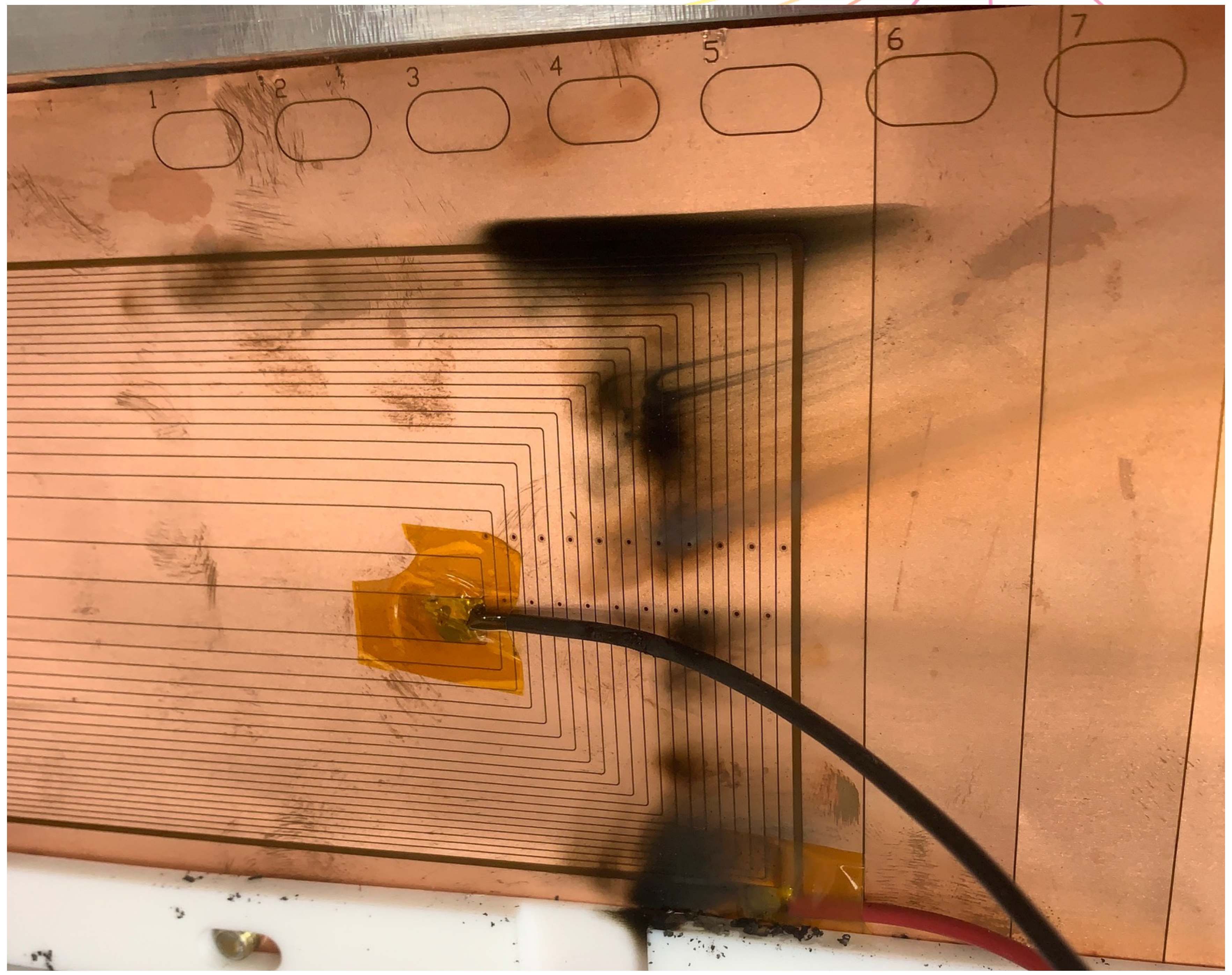
3-14 Å wavelength

signal = 500x IN11A

~ 50 t Cu

~ 0.6 MW max power, 50 kW average power

There were hiccups



High tech=breaks down

Lost cycle in June but we are back in business

It is Big !

From now on, we have the full

field integral 12ns @7Å 1.3ns @ 3.2Å

Full Q range

3.5Å^{-1} @ 3.2Å

Static structure factor of $Ca_{0.4}K_{0.6}(NO_3)_{1.4}$ (CKN)

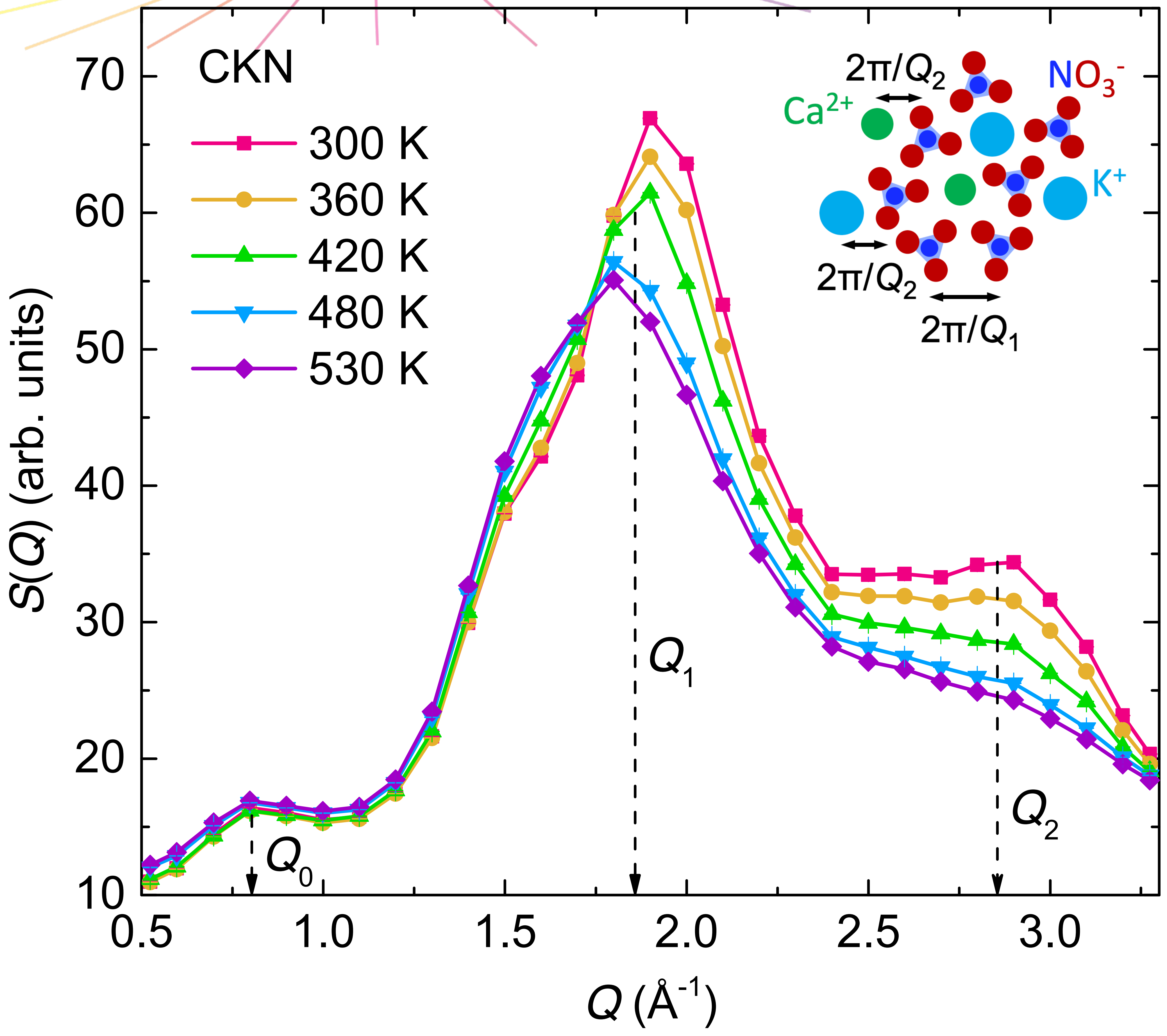
Melting point $T_m = 483$ K

Glass transition temperature $T_g = 333$ K

	σ_{coh} (barn)	σ_{inc} (barn)
N	11.01	0.5
O	4.232	0.0008
Ca	2.78	0.05
K	1.69	0.27

H. Senapati, et al., J. Phys. Chem. 95, 7050 (1991)
https://www.nist.gov/ncnr/planning-your-experiment/scattering-length-periodic-table

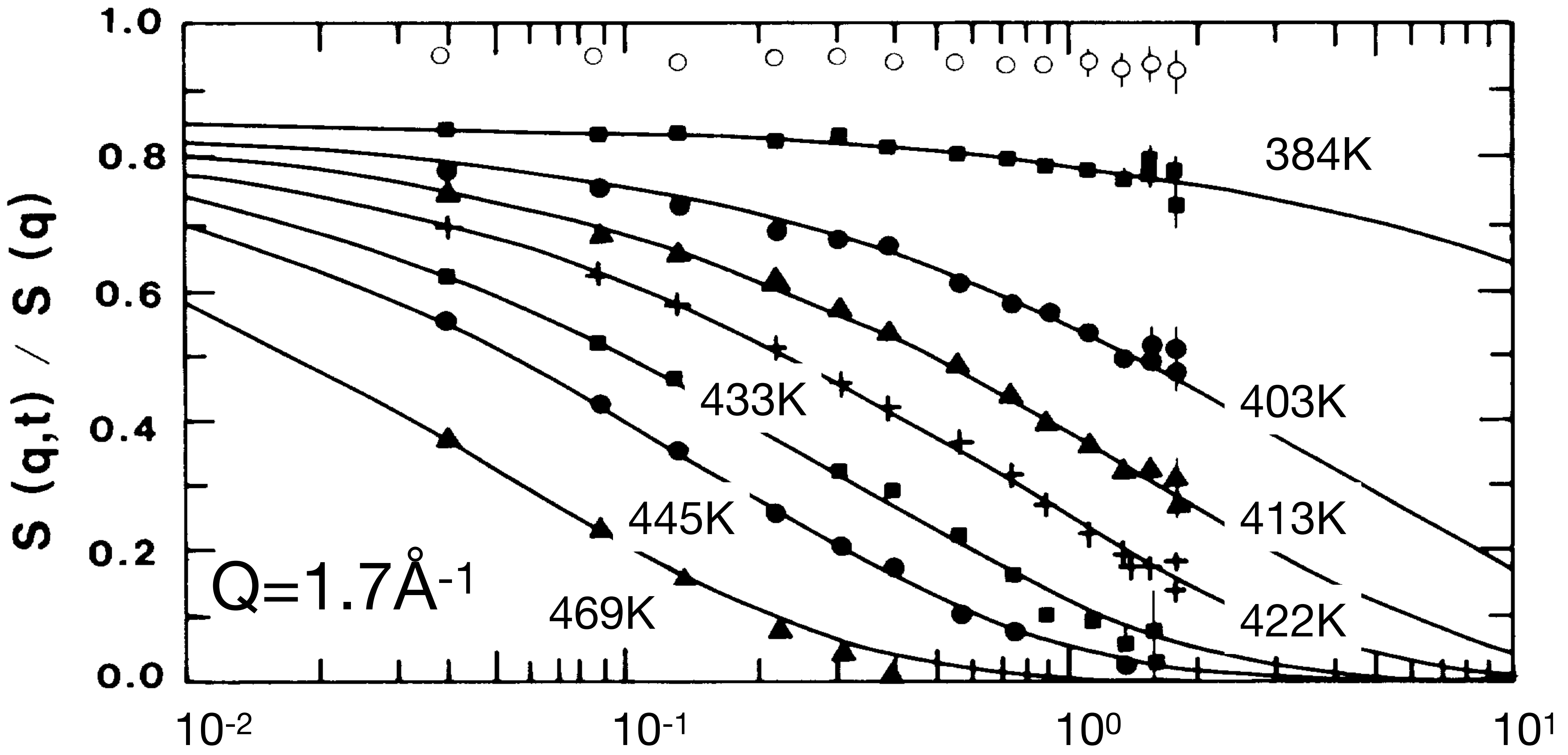
N and O dominates. Q_0: NO_3 chains
Q_1: NO_3 structure,
Q_2: NO_3 cation correlations



Measured on AMATERAS with $\lambda = 3.26$ Å

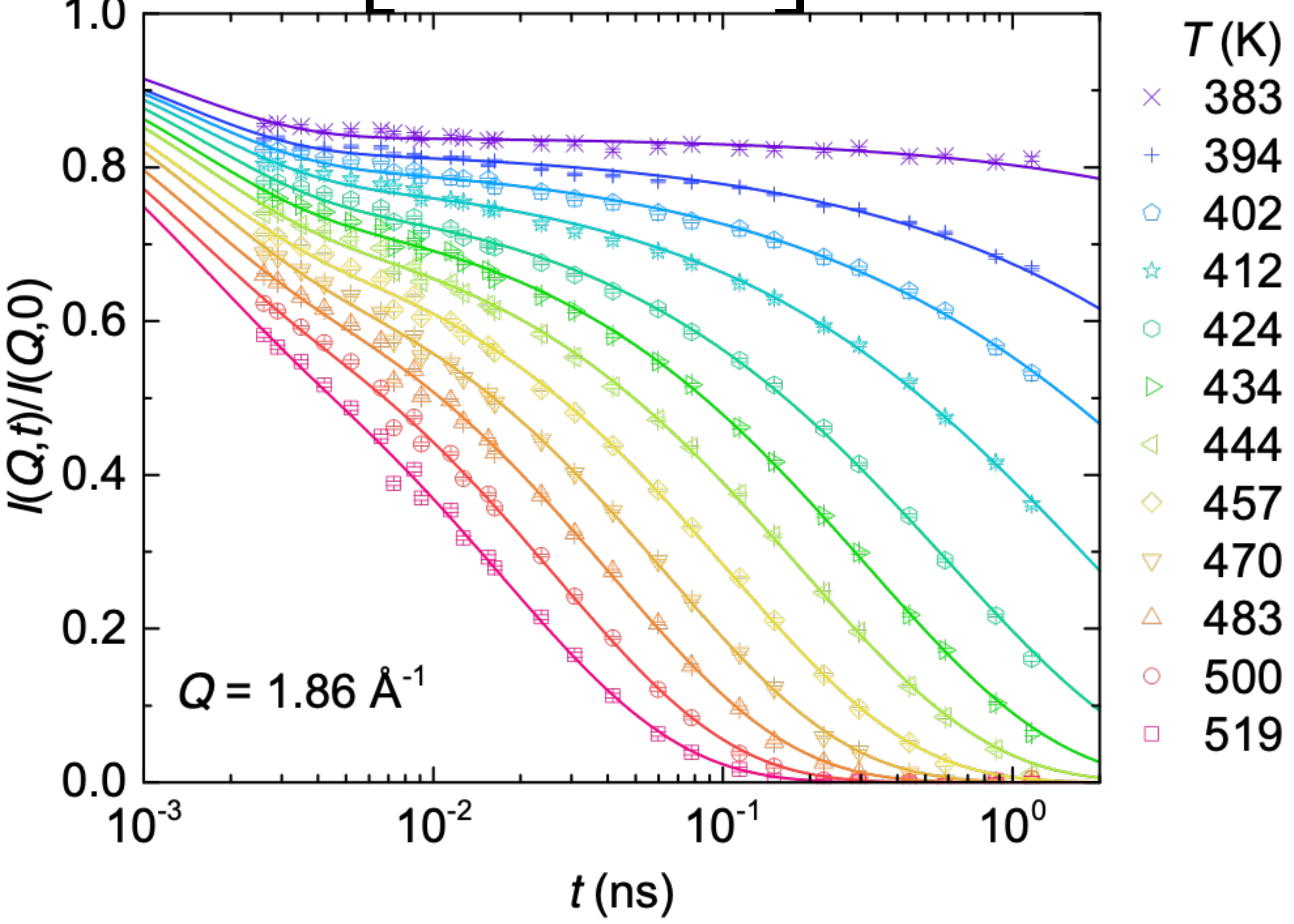
Intermediate scattering functions measured on WASP with λ = 4 Å

$$\frac{I(Q,\ t)}{I(Q,0)} = [1-f]\exp\left[-\left(\frac{t}{\tau_{\text{fast}}}\right)\right] + f\exp\left[-\left(\frac{t}{\tau_{\text{slow}}}\right)^{\beta}\right]$$



Neutron spin echo study of dynamic correlations near the liquid-glass transition

F. Mezei, W. Knaak, and B. Farago
Phys. Rev. Lett. 58, 571 – IN11. 1987.



Fast:local rearrangement Slow:long distance diffusion

P. Luo, …, Y Z*, ***Nature Communications*** 13, 2092 (2022)

Intermediate scattering functions measured on WASP with λ = 4 Å

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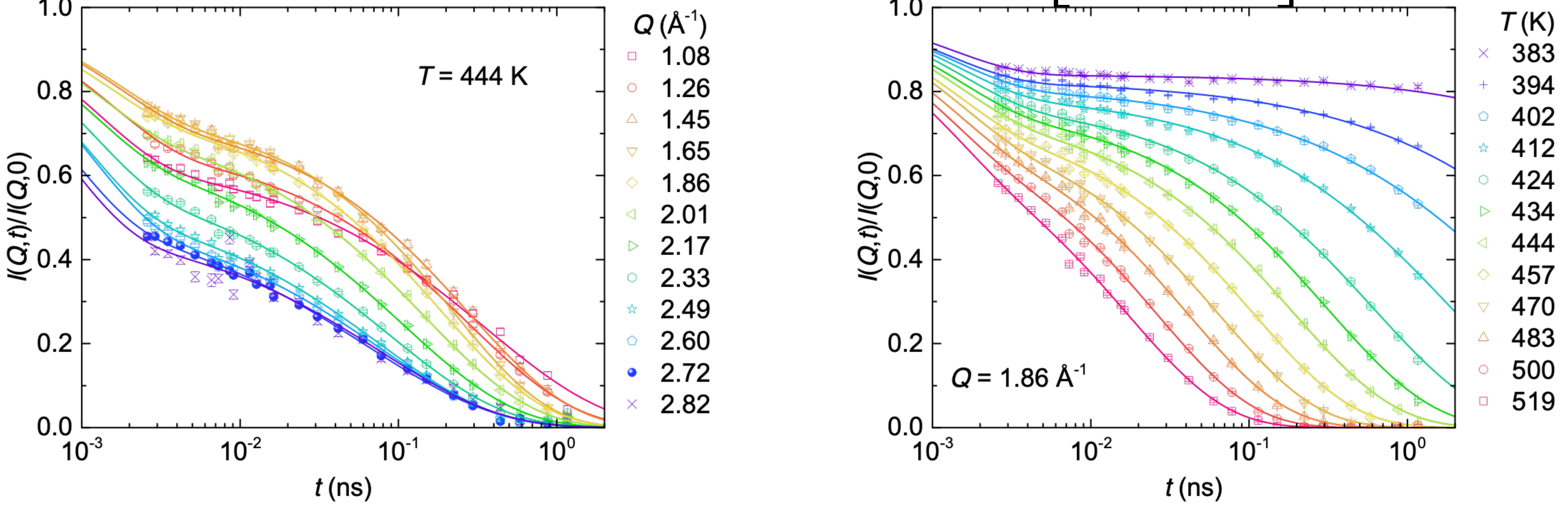
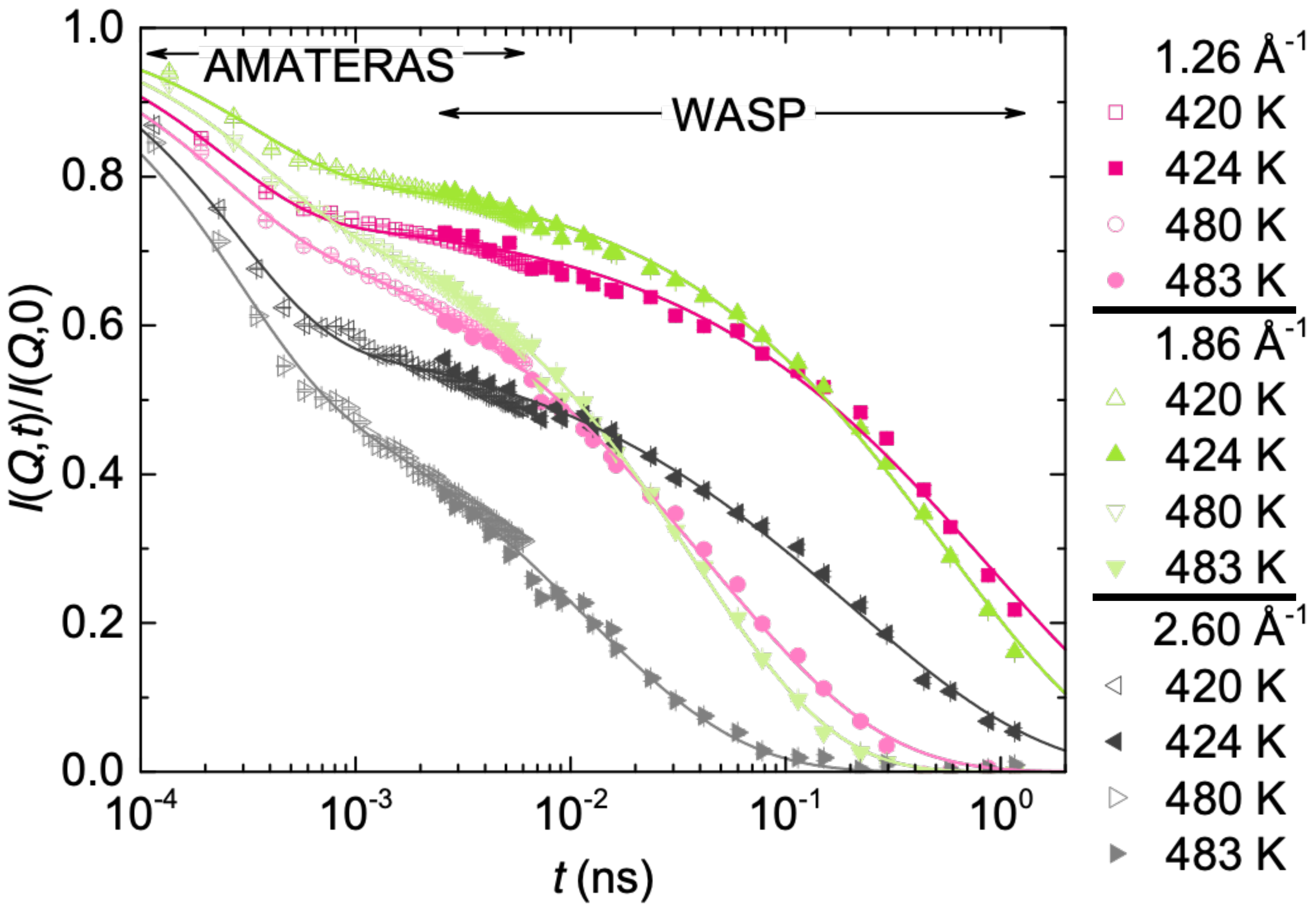


Fast:local rearrangement Slow:long distance diffusion

P. Luo, …, Y Z*, ***Nature Communications*** 13, 2092 (2022)

Joint ISFs of CKN covering a broad time window



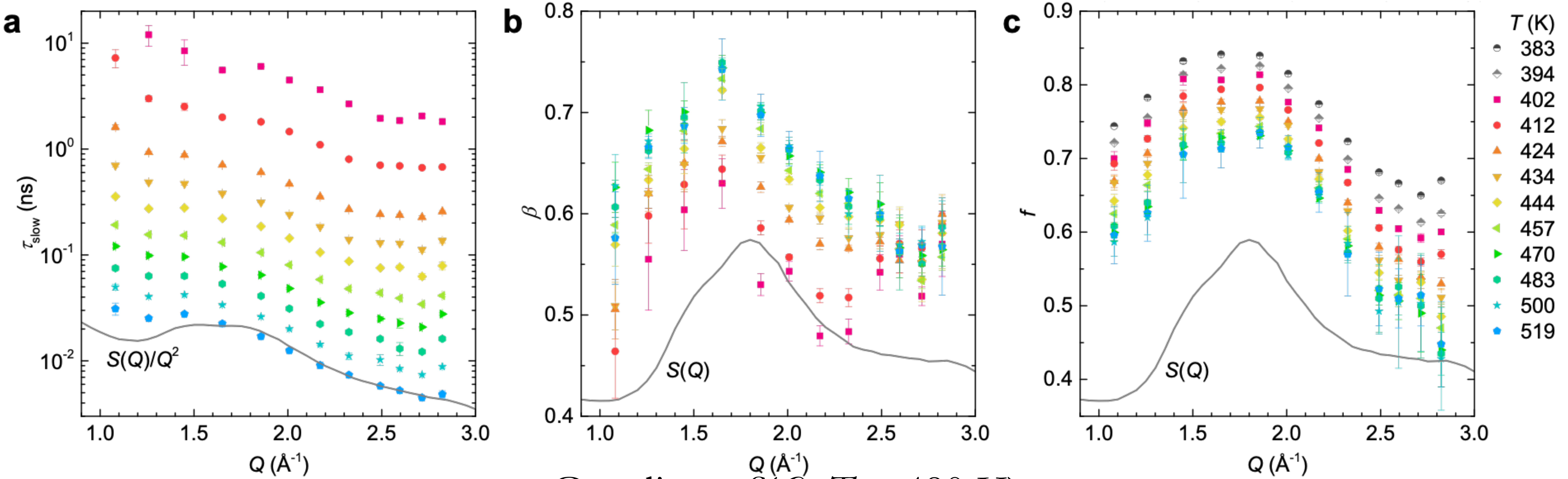
- TOF and WASP data match perfectly without any artificial adjustment of the data
- Confirms exponential line shape of the fast process

HIRES workshop

2023 Dec 12-15 @ ILL

P. Luo, …, Y Z*, ***Nature Communications*** 13, 2092 (2022)

Relaxation dynamics show strong Q-dependence of structure factor modulation

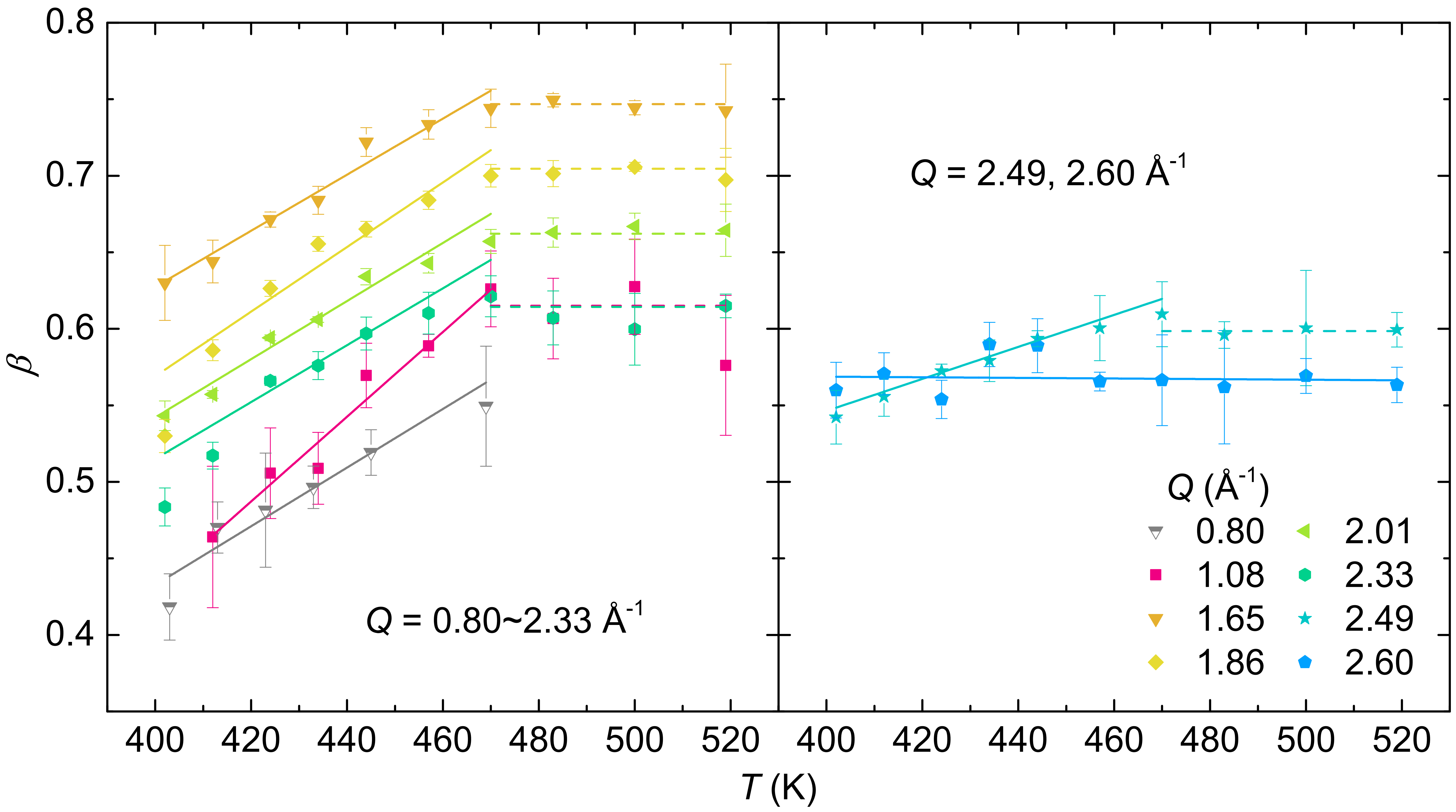


Gray lines: $S(Q, T = 480\ \mathrm{K})$

We recover the 1987 results at Q=1.7Å^{-1}. Controversy because prevailing MCT theories predict f=0.5 β=1. 300 citations!

P. Luo, …, Y Z*, ***Nature Communications*** 13, 2092 (2022)

Temperature dependence of the exponent β shows distinct behavior at Q > 2.4 Å^{-1}



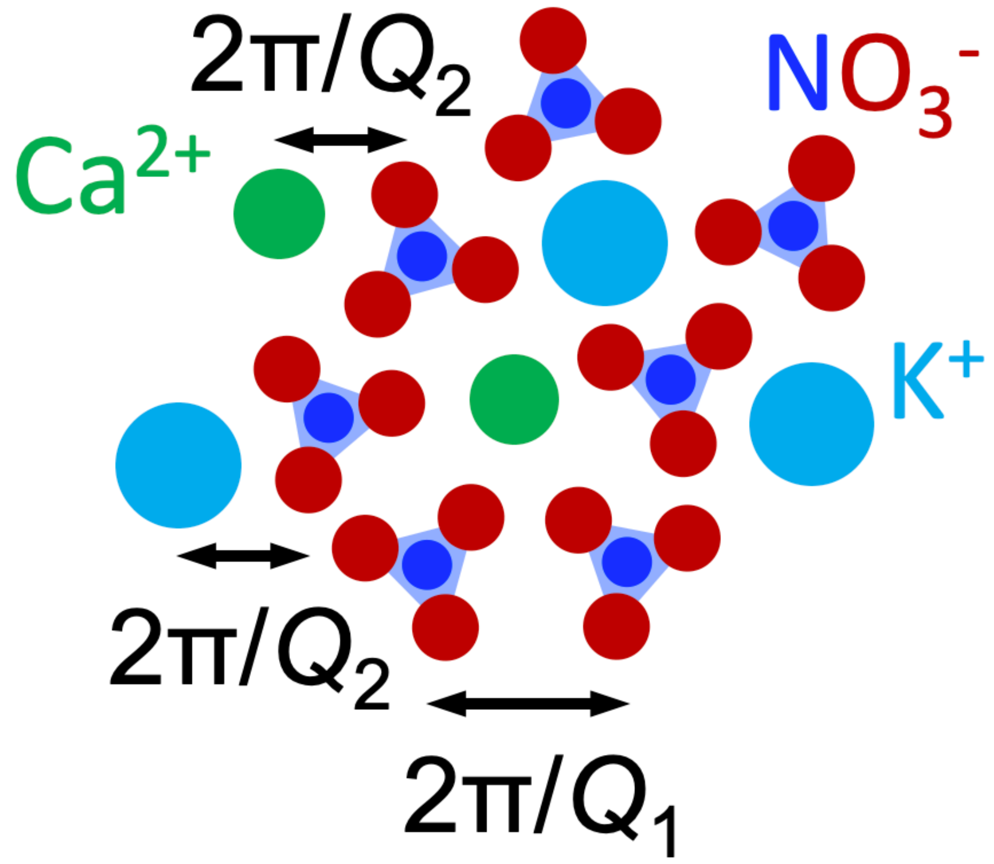
- For low viscosity liquid (T > 470 K), β is constant
- For supercooled liquid (T < 470 K):

i. At Q < 2.4 Å^{-1}, β decreases with decreasing temperature, suggesting increasing dynamic heterogeneity

ii. At Q > 2.4 Å^{-1}, β shows no temperature dependence + time constant has weaker temp. Dependence

P. Luo, …, Y Z*, ***Nature Communications*** 13, 2092 (2022)

CKN conclusion



1987 and 2022 results are consistent

<u>500x higher </u>intensity buys us:

- Wider time range more conclusive fits, Temperature dependence is clearly seen.
- Multiple q-values: dynamics strongly structure factor/Q dependent

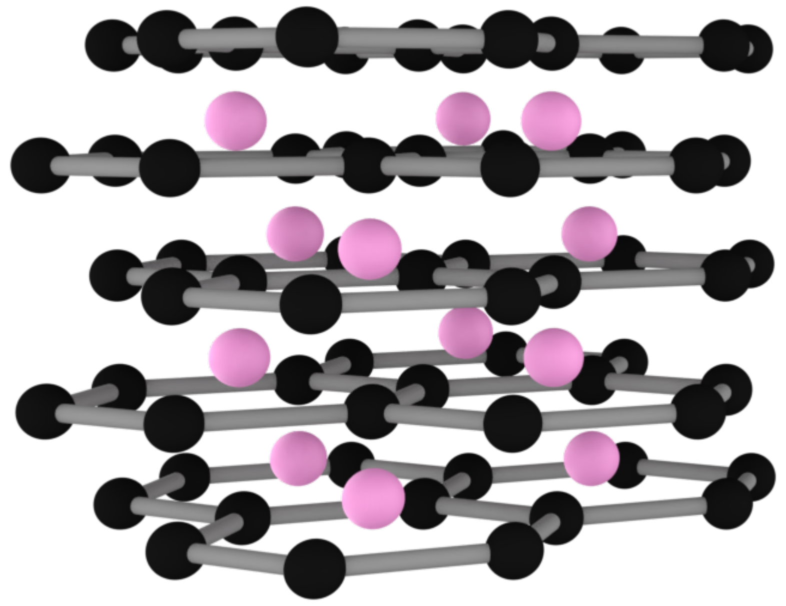
Beyond 2.4Å^{-1} the dynamics completely changes. Temperature dependence is much weaker due to the relatively stable Coulomb stabilised nitrate structure.

First student graduated

Diluted Li battery electrolyte dynamics

Filippa Lundin, prof. Aleksandar Matic
Chalmers University Göteborg

Ionic liquid based electrolyte, higher capacity by changing graphite anode to Lithium, non flammable BUT low conductivity



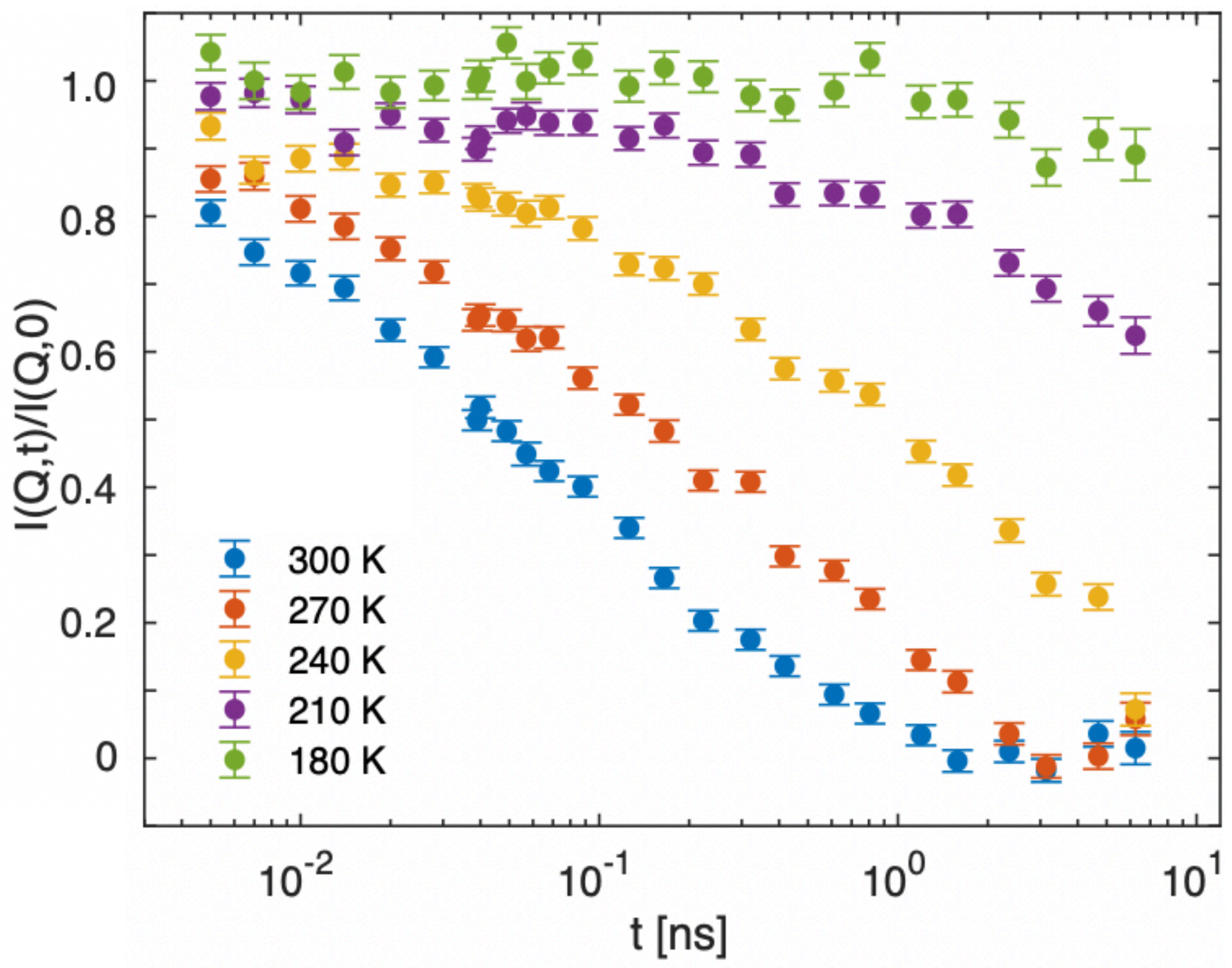
372 mAh/g

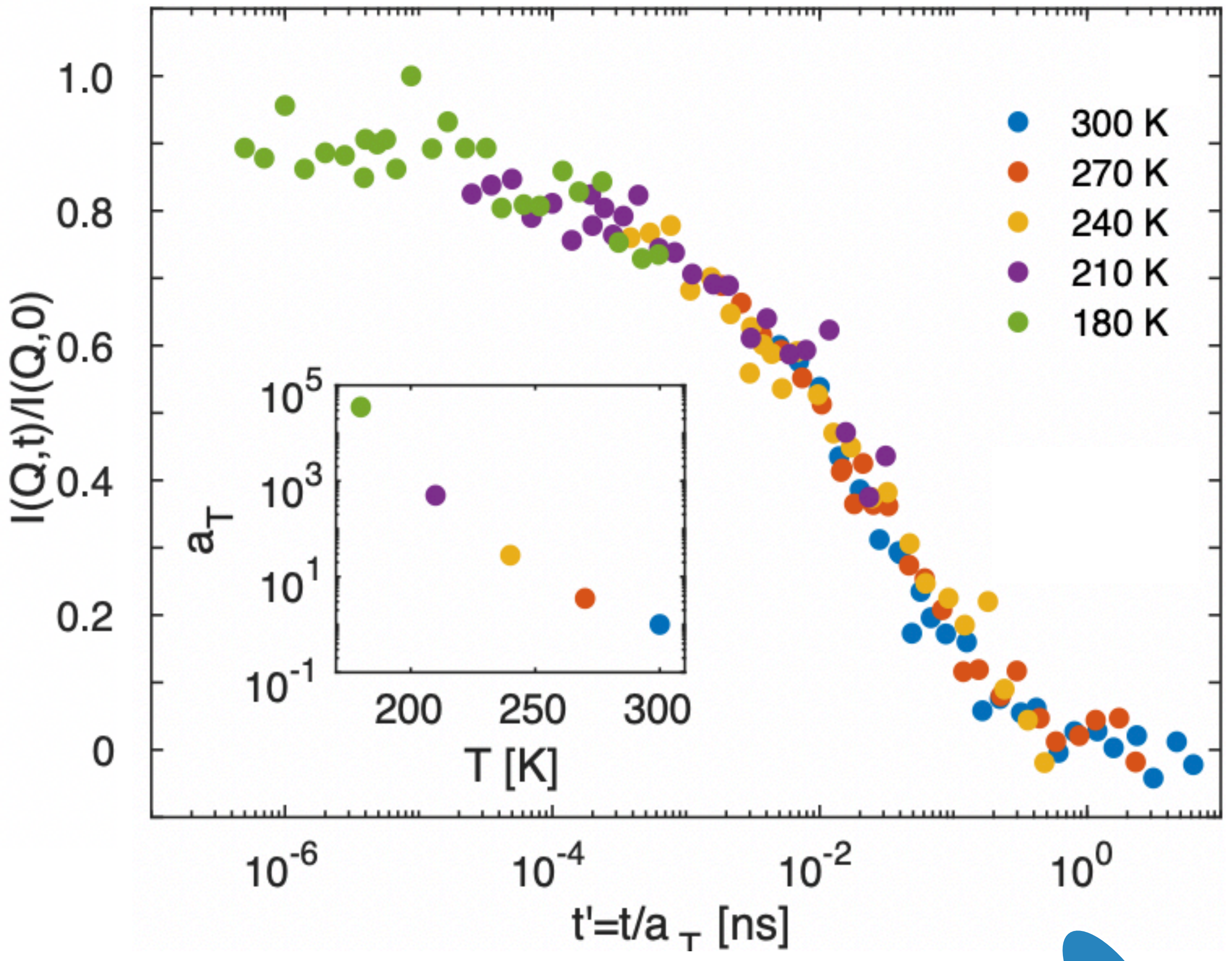
3862 mAh/g

Contradicting requirements, charge mobility, durability

Charge transport in diluted electrolyte

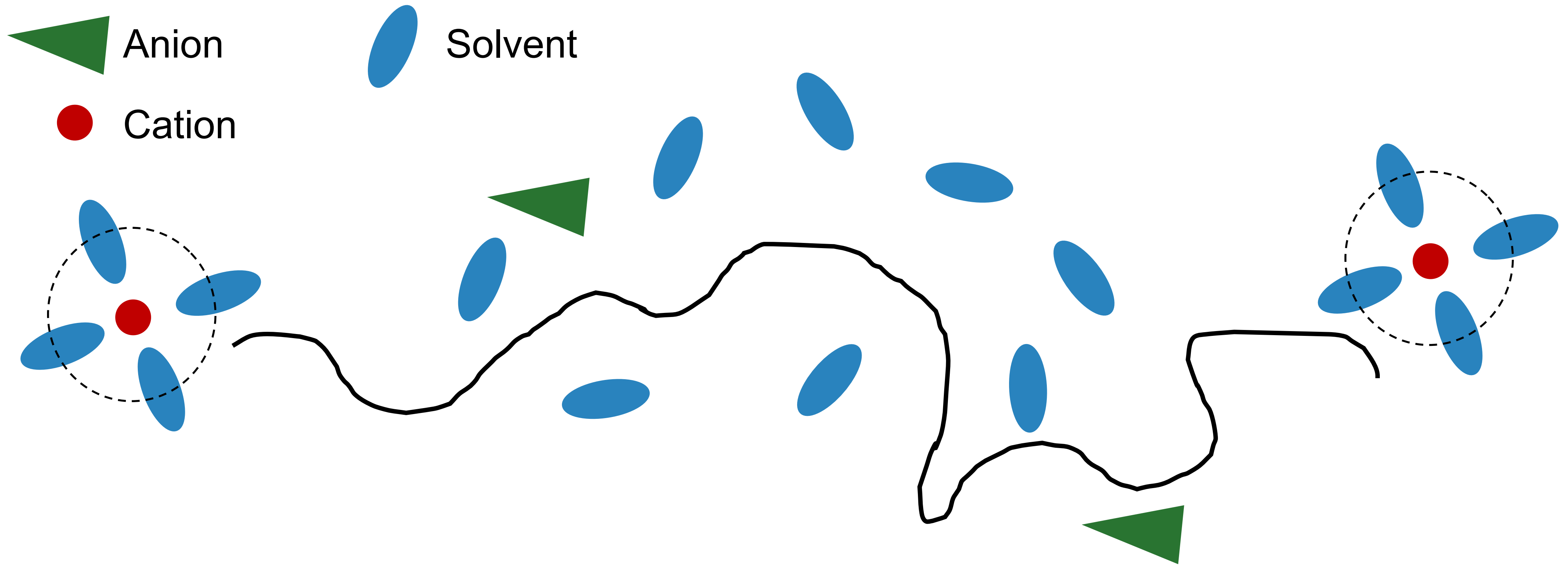
WASP covers full time range of motion

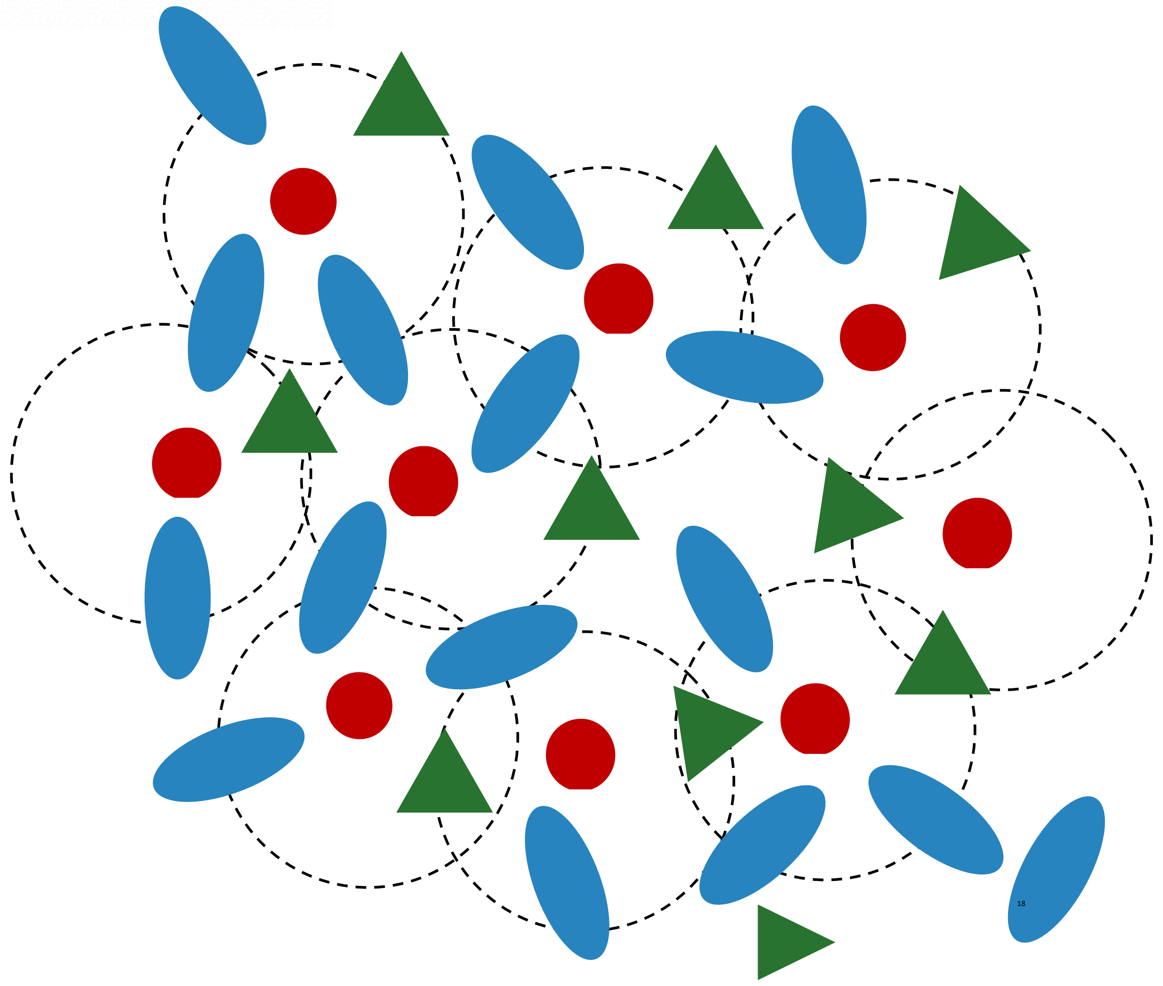




Low concentration: cation diffuses with solvation shell

High concentration: shells overlap; non-Gaussian diffusion

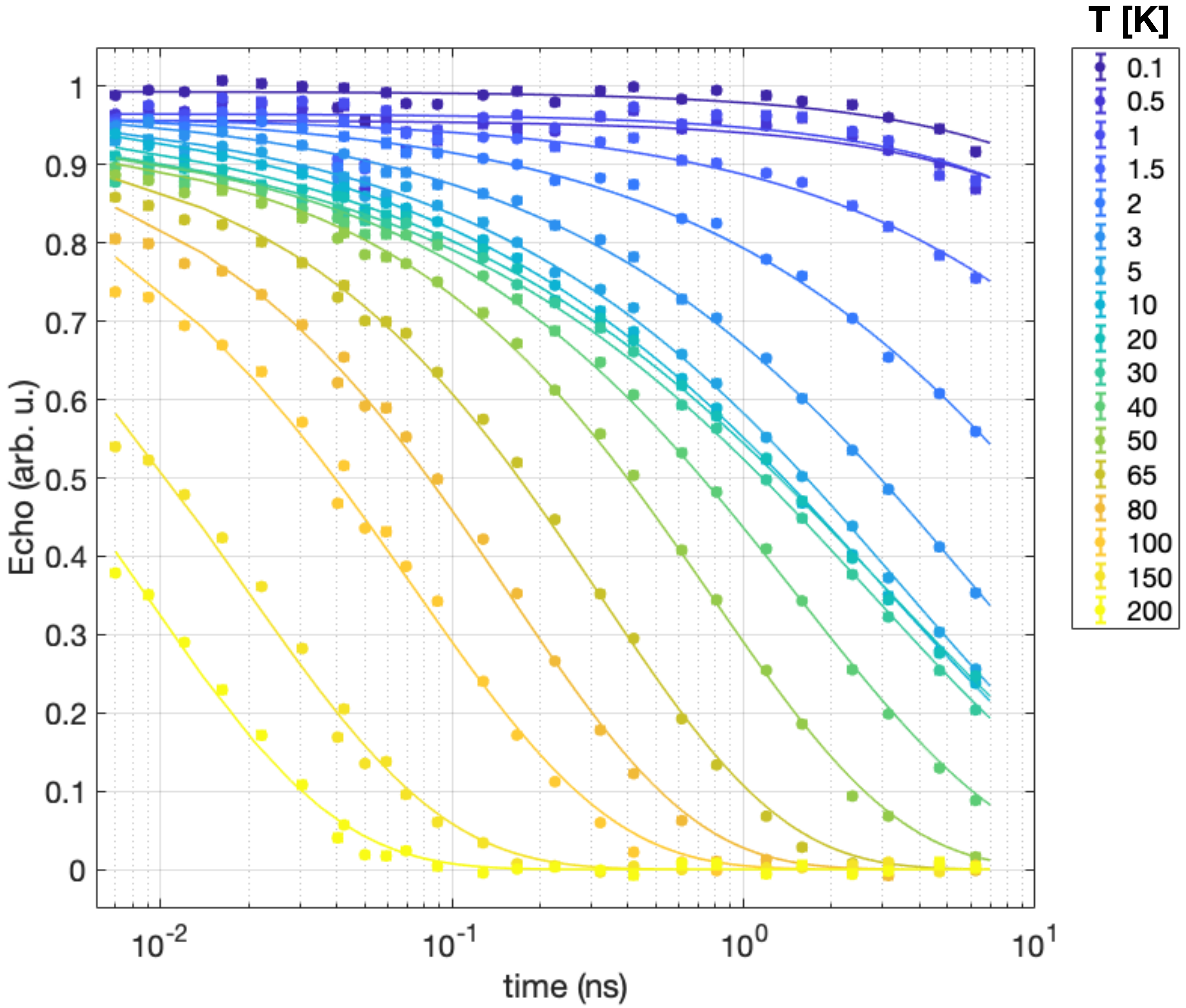


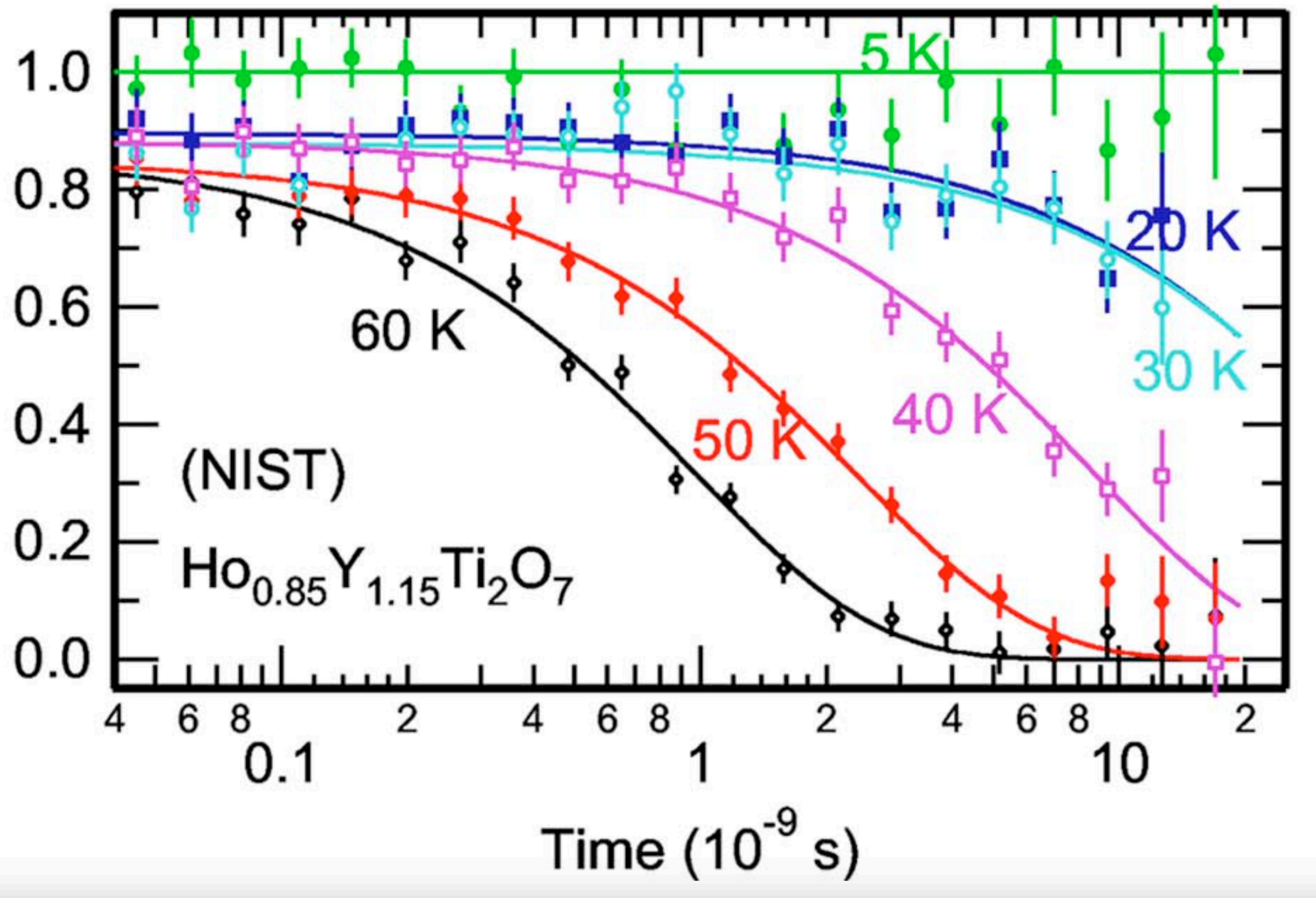


Right dilution conductivity improved, yet chemical compatibility kept, because Li solution shell not modified

J. Phys. Chem. C (2022), 126, 38, 16262–16271

Doped HoTiO spin glass





PHYSICAL REVIEW B 73, 174429 (2006)

When to use WASP ?

Our 3 weapons are:

High intensity

Wide time range (3-3.5 orders of magnitude)

Wide Q range

If you need:

Higher Q-range than high resolution NSE (IN15)

Longer time scales than BS, TOF

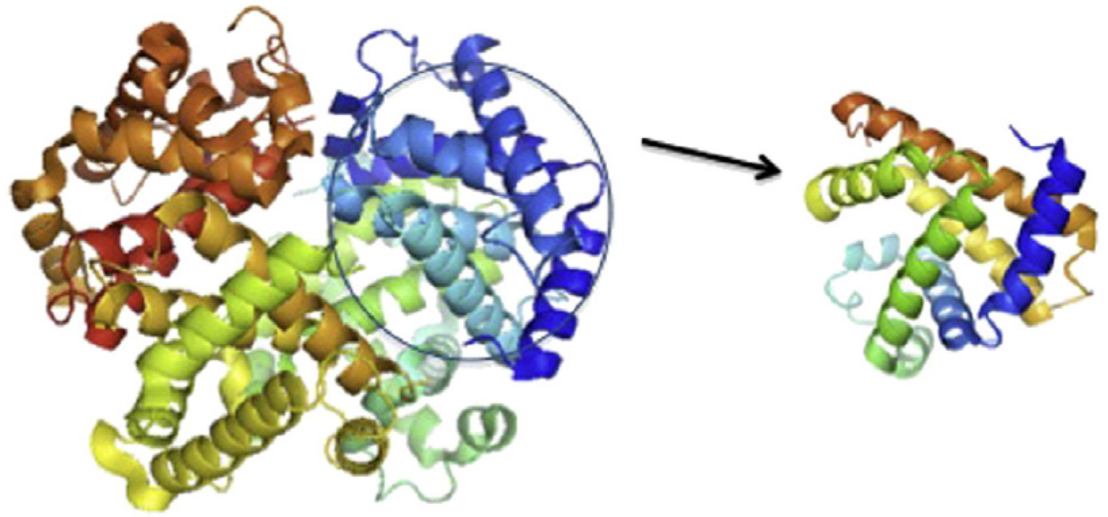
Polarised spectroscopy

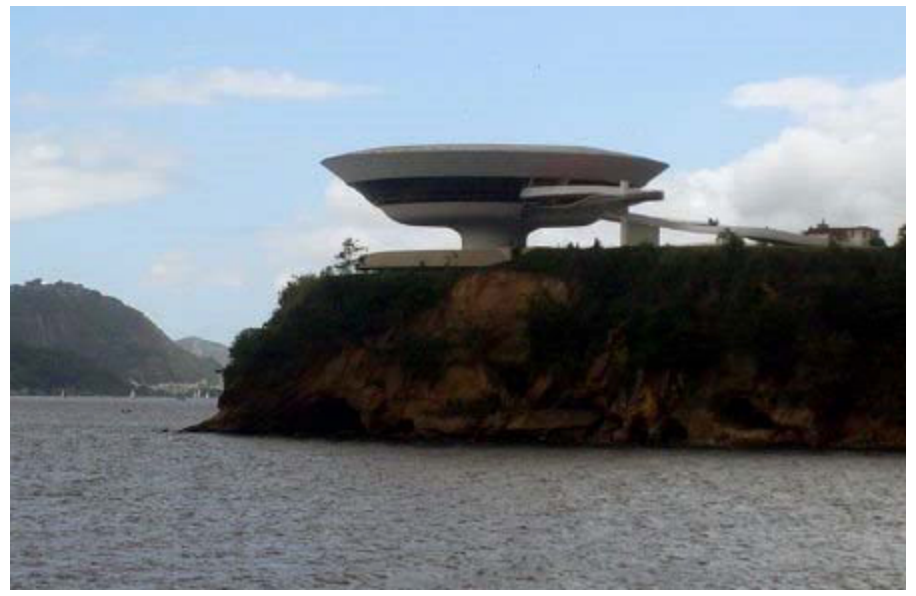
Why use on a pulsed source:

✦It works

✦Better monochromatization to resolve Bragg peaks

✦Many wavelength can be covered in one shot with massive detector coverage





Battery electrolytes
Fuel cells
Confined liquids
Ionic liquids
Spin ice,
Organic solar cells
Internal protein motions

Please register !

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DEADLINES

01/07	ABSTRACT submission
08/11	REGISTRATION

HIRES

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