



HIGH FLUX
ISOTOPE
REACTOR

SPALLATION
NEUTRON
SOURCE

Small-angle Neutron Scattering

Lisa DeBeer-Schmitt

NNS 2025



U.S. DEPARTMENT OF
ENERGY

ORNL IS MANAGED BY UT-BATTELLE LLC
FOR THE US DEPARTMENT OF ENERGY



A little about my career...



2008

Ph.D. at
University of
Notre Dame



2009 - 2010

Post-doc at
GP-SANS



2010 - 2011

Team Lead for SNS
Sample Environment



2011 - 2015

Scientific Associate
for Sequoia

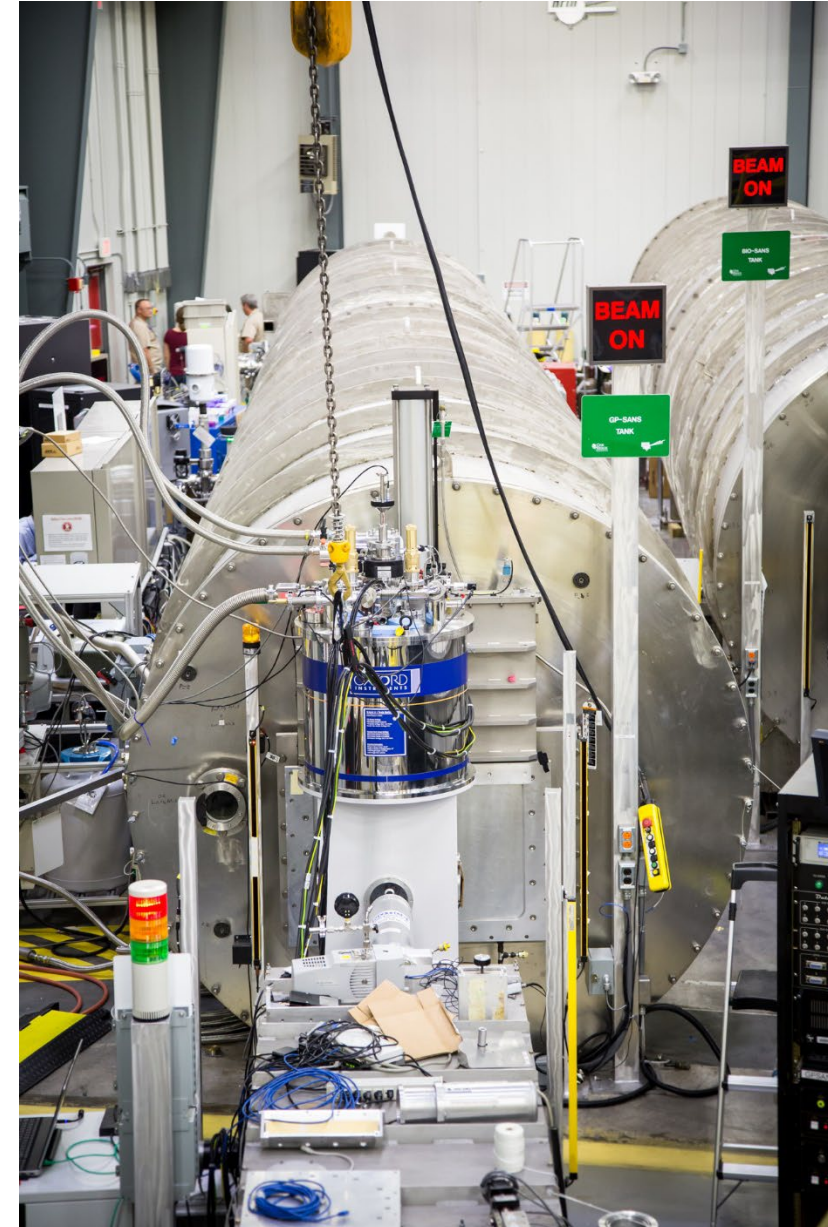


2015 - current

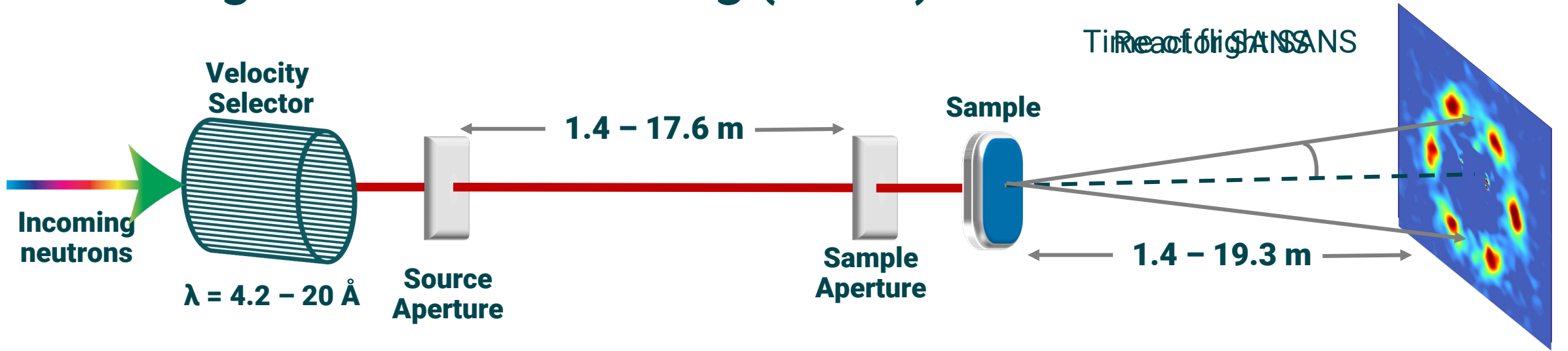
Point-of-contact
for GP-SANS

Outline:

- What is SANS?
- Instruments at ORNL
- Sample Environment capabilities
- Science Cases
 - Polymers
 - Magnetic ordering (diffraction)
 - Engineering materials
- New Directions



Small-angle neutron scattering (SANS)



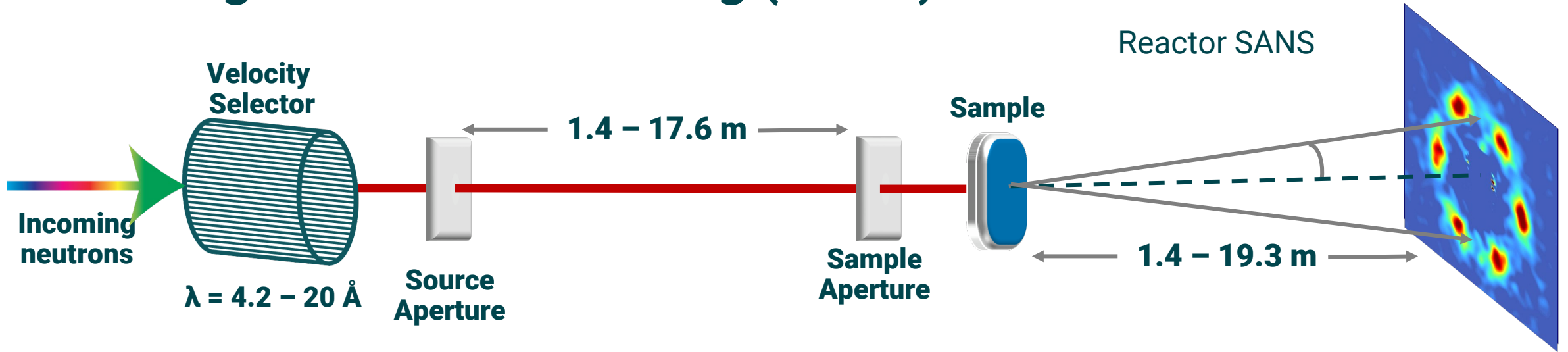
$$I(Q) = A \Delta\rho^2 n V^2 P(Q) S(Q)$$

Labels for the equation components:

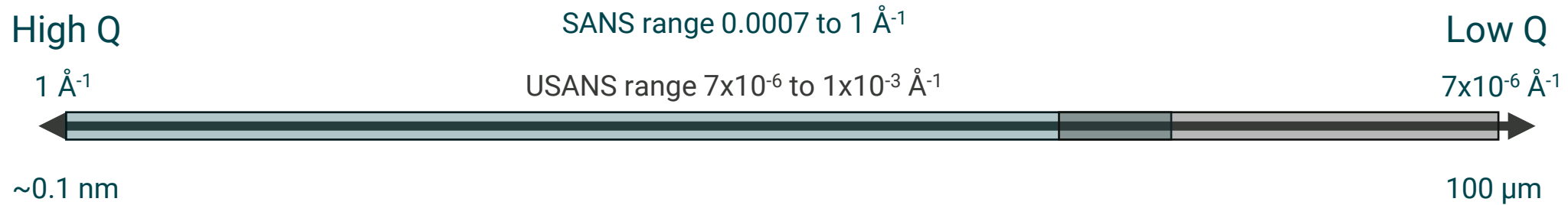
- A : Calibration
- $\Delta\rho^2$: Scattering length density
- n : Number of scatters
- V^2 : Volume fraction
- $P(Q)$: Form and structure factors
- $S(Q)$: Form and structure factors

Probes lengths of $0.5 - 700 \text{ nm}$
Using angles of $\theta = <0.1$ to 45°

Small-angle neutron scattering (SANS)

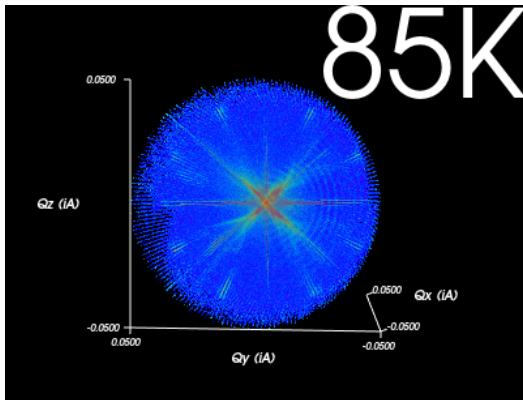


Q vs Real-space Length Scale in SANS



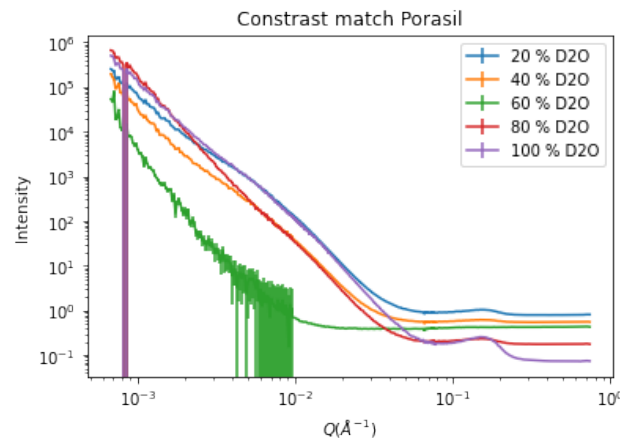
Why Neutrons?

- Low Energy ($E=80\text{meV}$ for 1 \AA neutrons vs. 12.42 keV for x-rays. **No radiation damage.**)
- High Penetration of cold neutrons – bulk samples
- Large difference in the scattering cross-section for the hydrogen and deuterium- contrast variation capability.
 - Solute or solvent can be deuterated to vary the contrast ($\rho_{\text{H}_2\text{O}} = -0.56 \times 10^{10} \text{cm}^{-2}$, $\rho_{\text{D}_2\text{O}} = 6.334 \times 10^{10} \text{cm}^{-2}$)
 - Study of multicomponent systems through selective deuteration and contrast matching with H/D mixtures.
- Sensitive to substantial difference in scattering length density in transition metals
- **MAGNETISM.**
 - Neutron has a magnetic moment and spin which give contrast

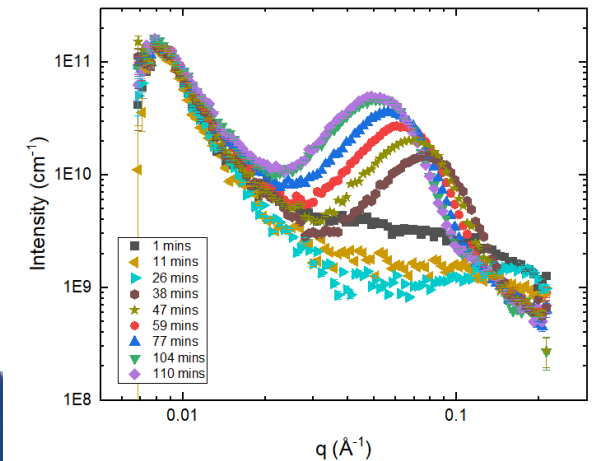
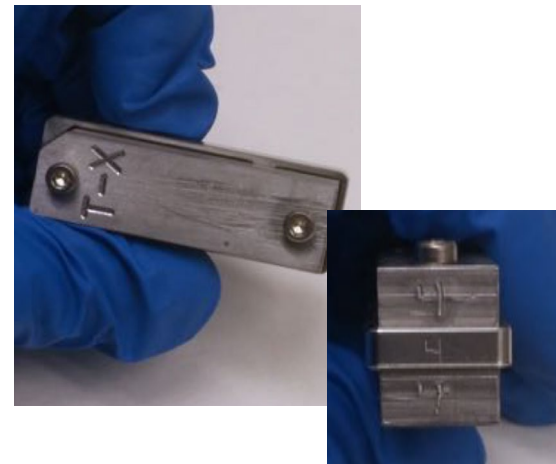


Magnetic domains in FeV_2O_4

L. Kish et al in preparation



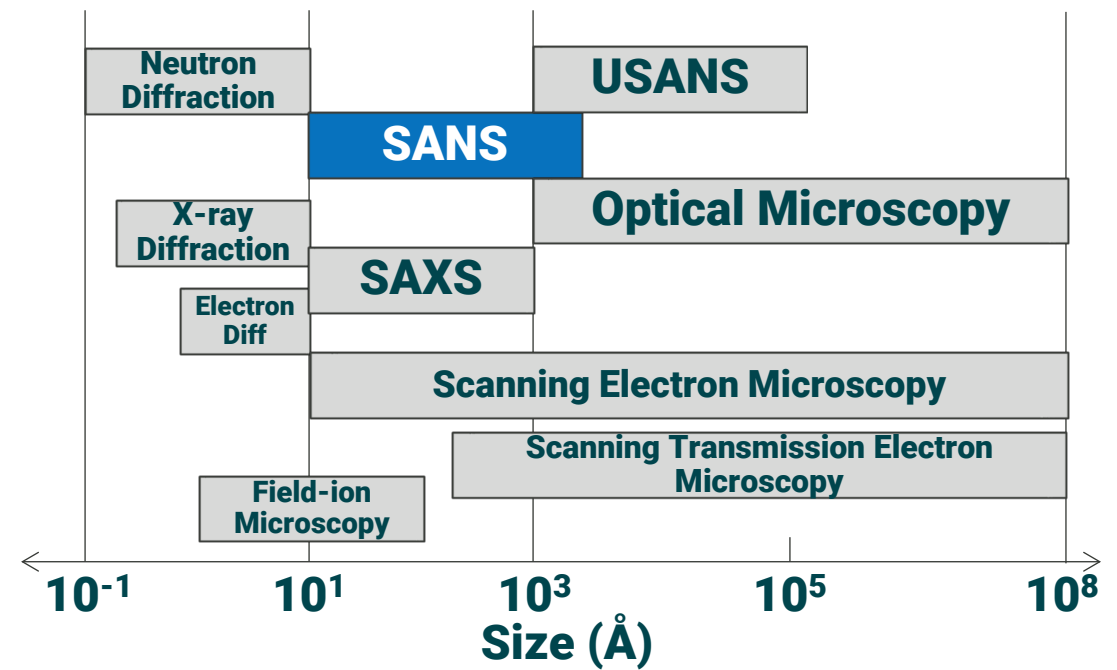
Data from NXschool experiment



Copper precipitate growth in steel

SANS with other techniques

- Microscopy: Direct but limited



- SAS: Indirect and model-dependent but in-situ

SANS with other techniques

- Microscopy: Direct but limited



Finally discovered in 1981 and a second in 2003

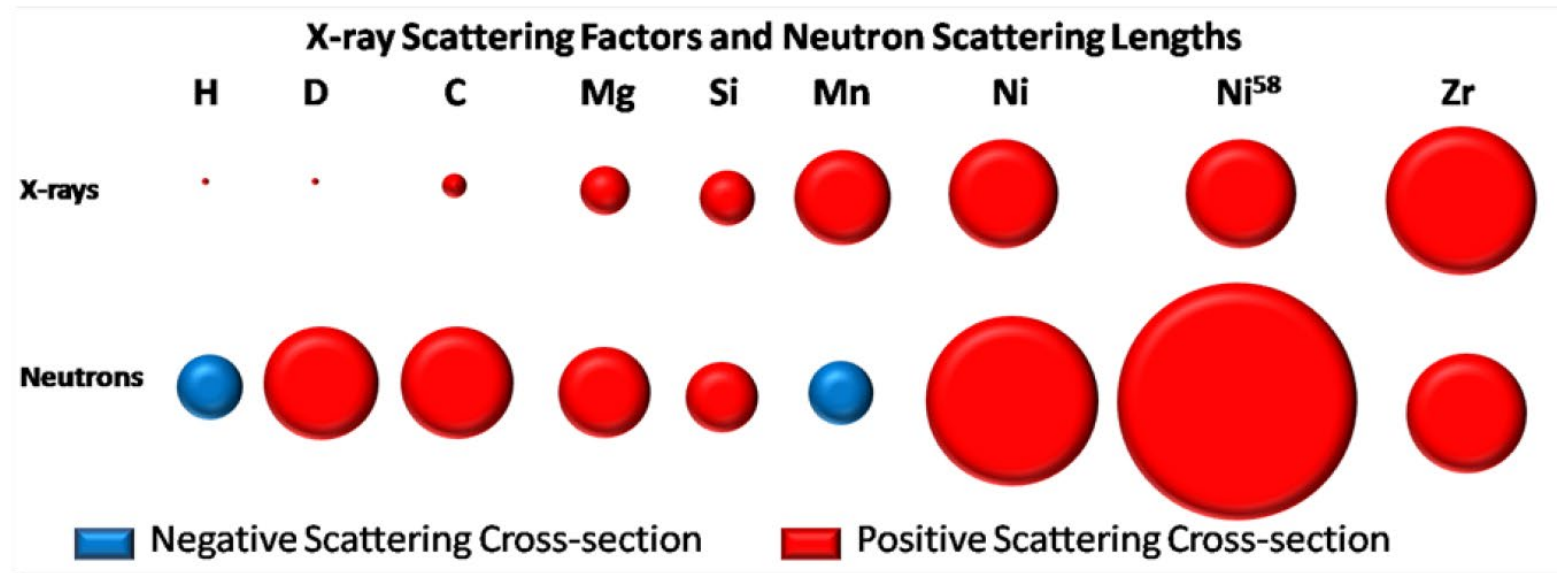


- SAS: Indirect and model-dependent but in-situ

Neutron and x-ray scattering cross-sections

X-ray and neutron scattering are essentially the same concept, except...

- X-rays scatter from electrons

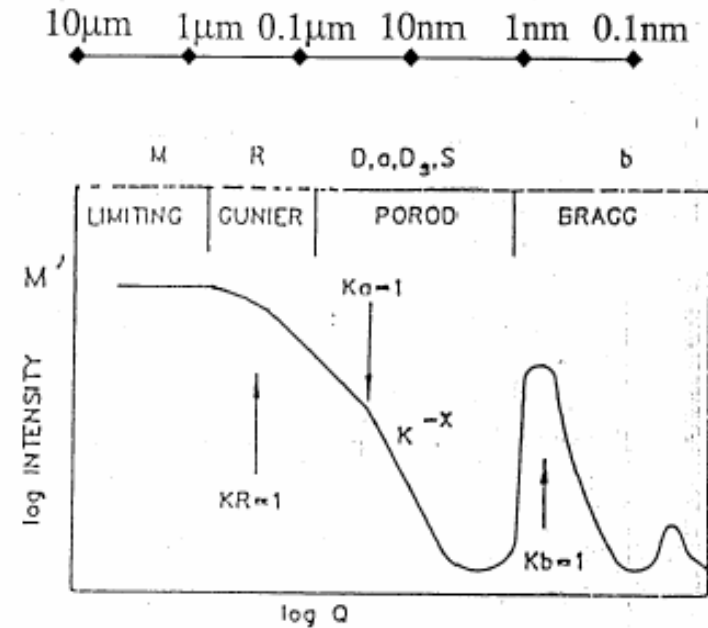
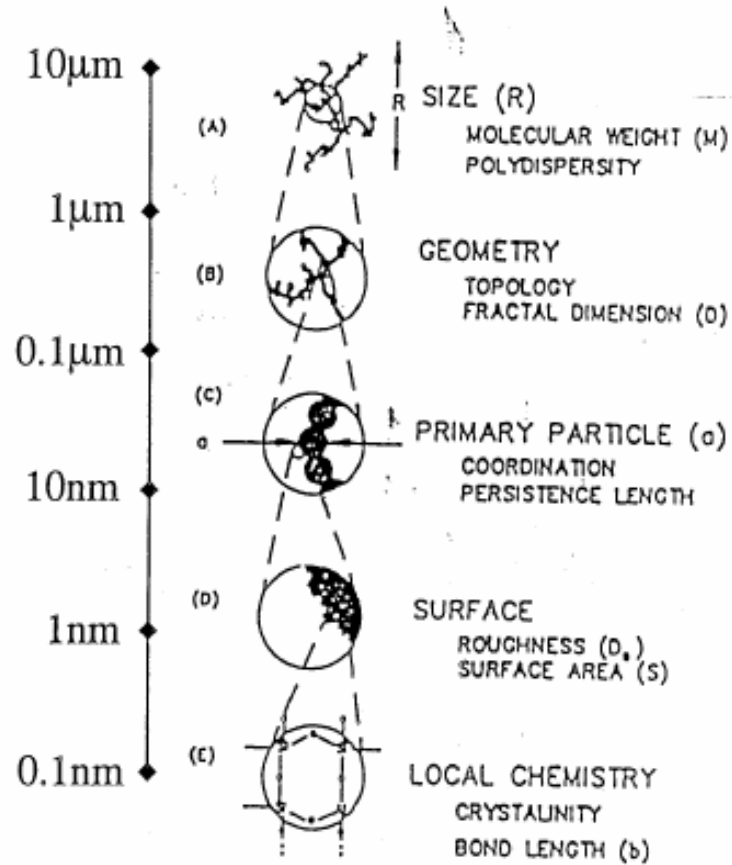


<http://www.ncnr.nist.gov/resources/n-lengths/>

http://www.isis.rl.ac.uk/ISISPublic/reference/Xray_scattfac.htm

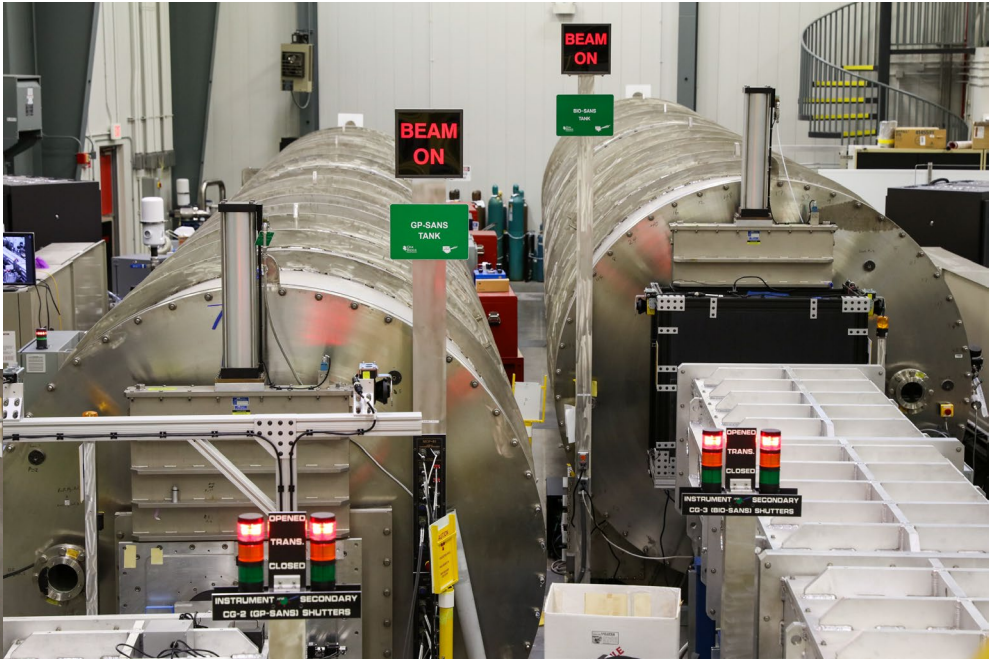
C. Metting, Dissertation University of Maryland 2011

SANS from hierarchical structures



SANS is a structural technique that probes a wide array of length scales

SANS Suite



ORNL SANS instrument specifications.

	GP-SANS	Bio-SANS	EQ-SANS	USANS
Moderator	Supercritical hydrogen	Supercritical hydrogen	Coupled, supercritical hydrogen	Decoupled, poisoned hydrogen
Source	Reactor, continuous	Reactor, continuous	Spallation, 60 or 30 Hz, TOF	Spallation, 60 Hz, TOF
Sample-to-detector distance	1.1–20 m	1.1–15.5 m	1.0–9 m	N/A
Beam size	Up to 20 mm diameter	Up to 20 mm diameter	Up to 15 mm diameter	Up to 40 mm by 40 mm
Q range	7×10^{-4} to 1 \AA^{-1}	9×10^{-4} to 1 \AA^{-1}	2×10^{-3} to 6 \AA^{-1}	7×10^{-6} to $5 \times 10^{-3} \text{ \AA}^{-1}$
Incident wavelength	4–25 \AA	6–25 \AA	0.5–20 \AA	3.6, 1.8, 1.2 and 0.9 \AA
$\Delta\lambda/\lambda$	9–45%	9–45%	<1–20%†	Monochromated with crystals
Detector type	^3He LPSD array	^3He LPSD array	^3He LPSD array	^3He

† The resolution of EQ-SANS is wavelength dependent and also depends on the binning used in the software. The upper end of the range indicated is for the shortest wavelength used in the current data reduction.

Sample Environments:

Current Sample environment options:

- New Robot sample changer
 - Peltier system that goes from -10 to 120 °C with 0.01 degree control and holds 42 samples at temperature
 - Atmosphere Furnace up to 300 °C
- Rheometer
- New controlled atmosphere furnace and vacuum furnace to > 1000 °C
- Horizontal 11 T recondensing magnet allowing users to utilize 11 T at 30 mK. (3/2016), 5 T horizontal open bore magnet (5/2017) and 8 T vertical magnet with temps 30 mK to 300 K
 - Electric field and current can also be applied to the samples
- SANS dedicated cryostat with sapphire windows
- Pressure cells (McHugh 2kBar and 1kBar changer)

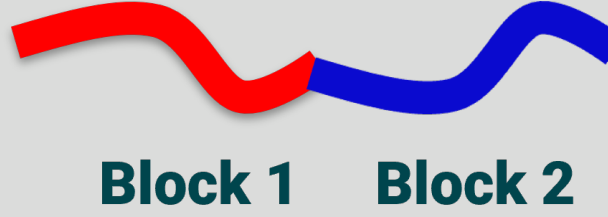
Future upgrades:

- Strain cell to apply in-situ compression or tension (11 T and 5 T magnets)
- In-situ reaction vessel
- Load frame for metallurgical specimens

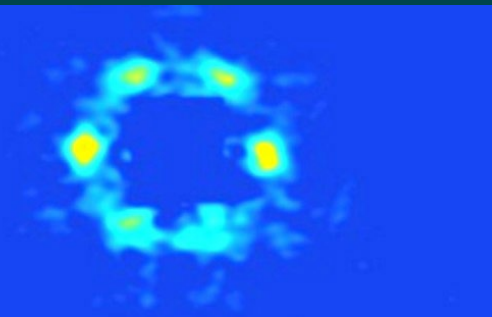


Science cases

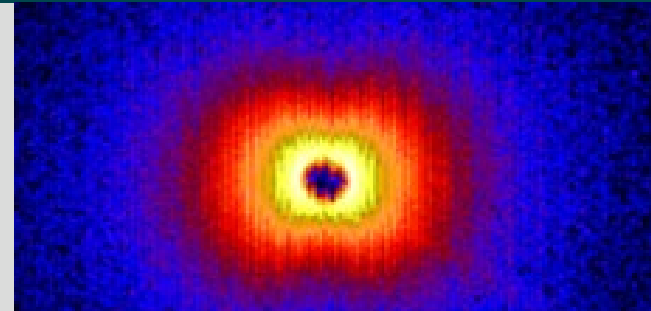
Diblock
copolymers



Skyrmion in
thin films



Novel Steels
for reactor



Influence of Cleavage of Photosensitive Group on Micellization and Gelation of a Doubly Responsive Diblock Copolymer

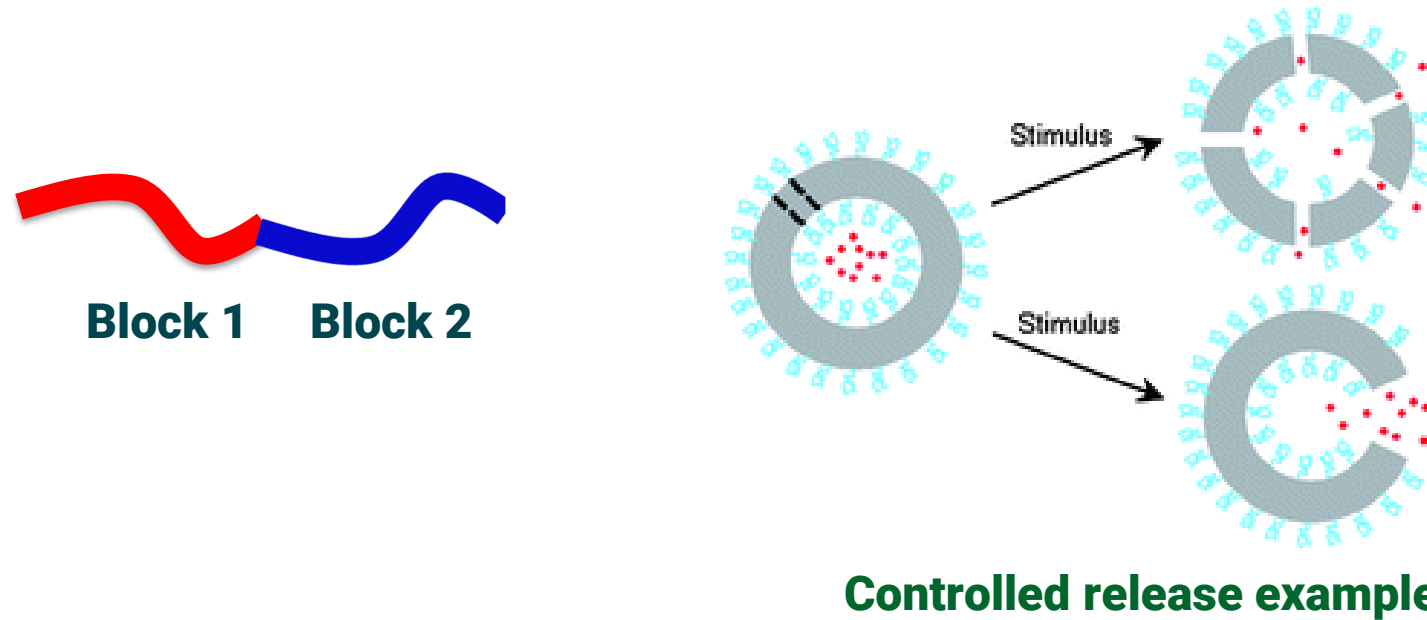
Lilin He¹, Bin Hu², Daniel M. Henn², and Bin Zhao²

¹ Large Scale Structure Group, Neutron Scattering Division, Oak Ridge National Laboratory

² Department of Chemistry, University of Tennessee: Knoxville

Influence of Cleavage of Photosensitive Group on Micellization and Gelation of a Doubly Responsive Diblock Copolymer

Block Copolymers have wide technical applications owing to their capabilities to self-assemble into nanostructures under different conditions.



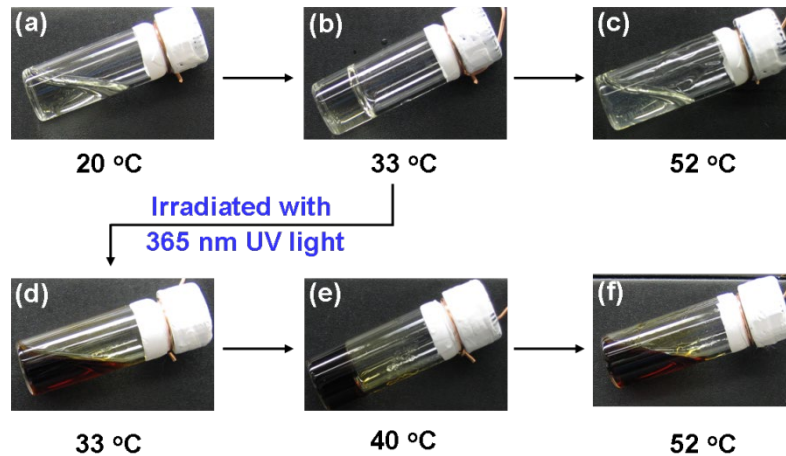
Challenge: To precisely **tune** and **control** the molecular characteristics of the block copolymers under appropriate conditions.

Thermal- and light-sensitive diblock copolymer PEO-b-P(TEGEA-co-NBA)

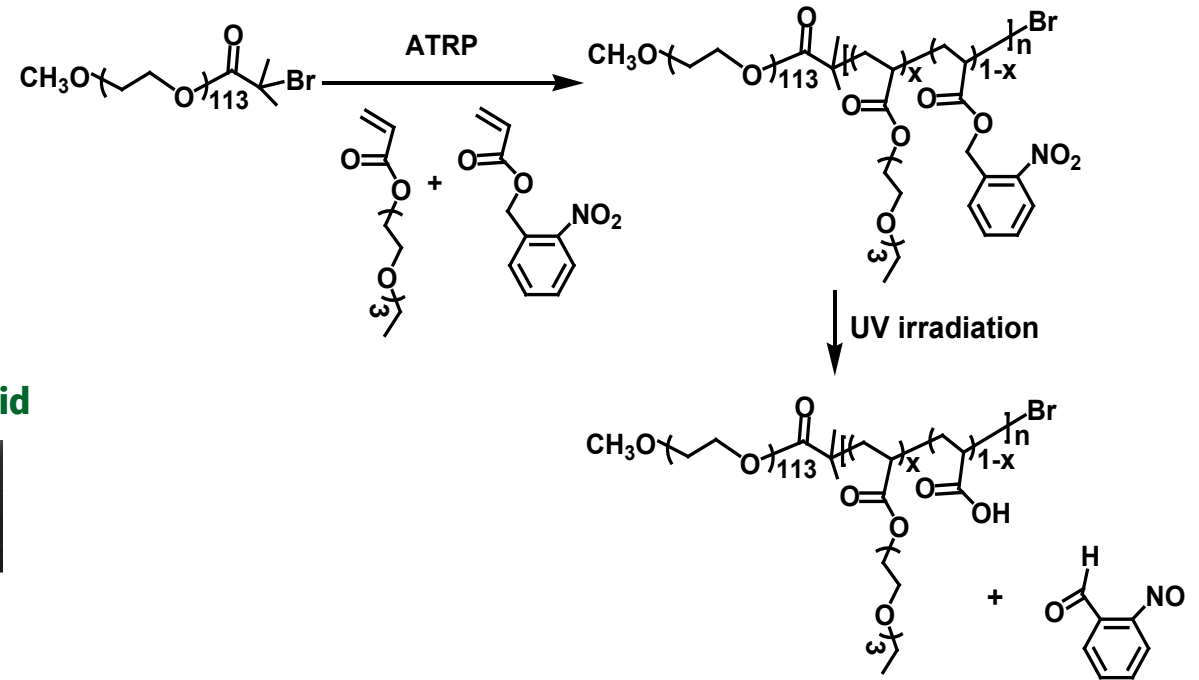


Prof. Bin Zhao, UTK

Flowing Fluid Gel Flowing Fluid



- **Brown-red color due to the photochemical reaction**
- **Multiple transitions**

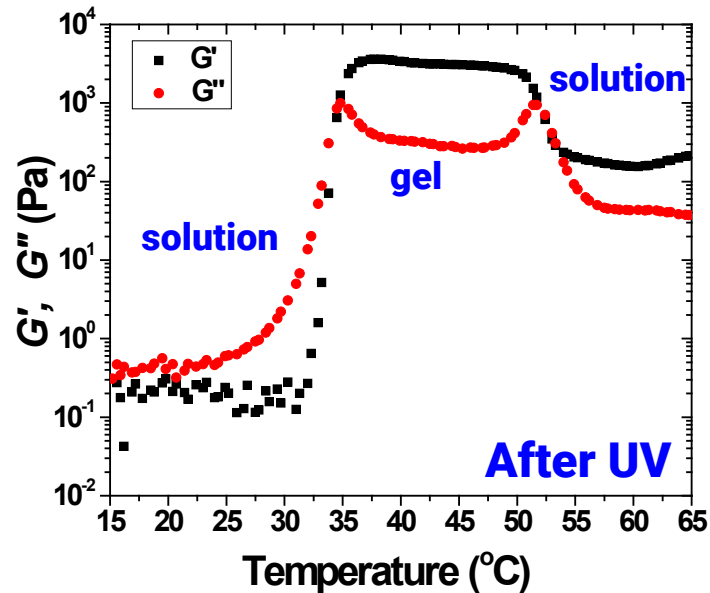
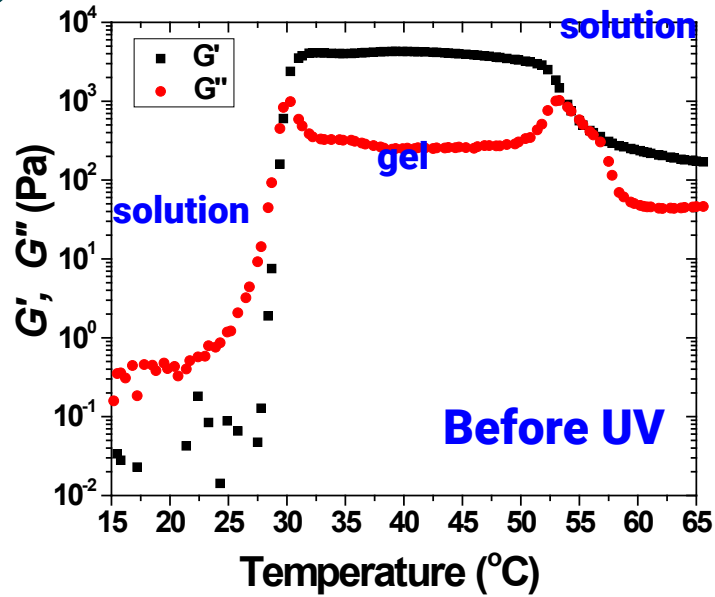


- **“living”/controlled radical polymerization**
- **Irradiated for 6 days with 365nm UV light**



Block 1 Block 2

Rheological Results



- Dynamic storage modulus G' , loss modulus G'' , versus temperature
- 25wt % D₂O solution of PEO-*b*-P(TEGEA-*co*-NBA).
- A heating rate of 3°C/min.
- A strain amplitude of 1 %
- An oscillation frequency of 1 Hz.

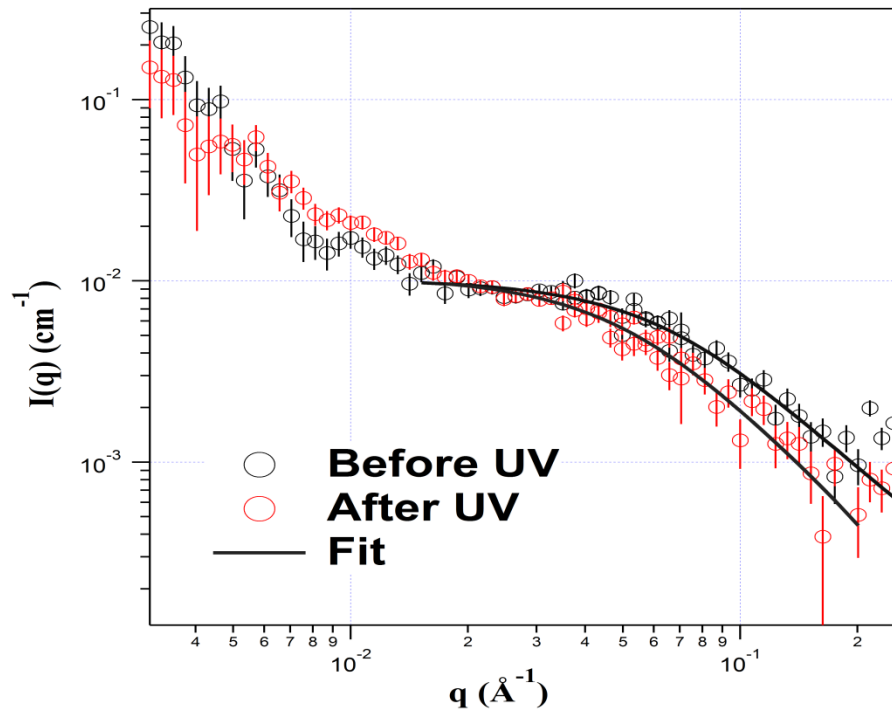
- Reversible sol-gel-soft gel transitions is achieved;
- Gel is composed of packed micelles;
- UV irradiation leads to a narrower gel state window.

Rheological
Properties

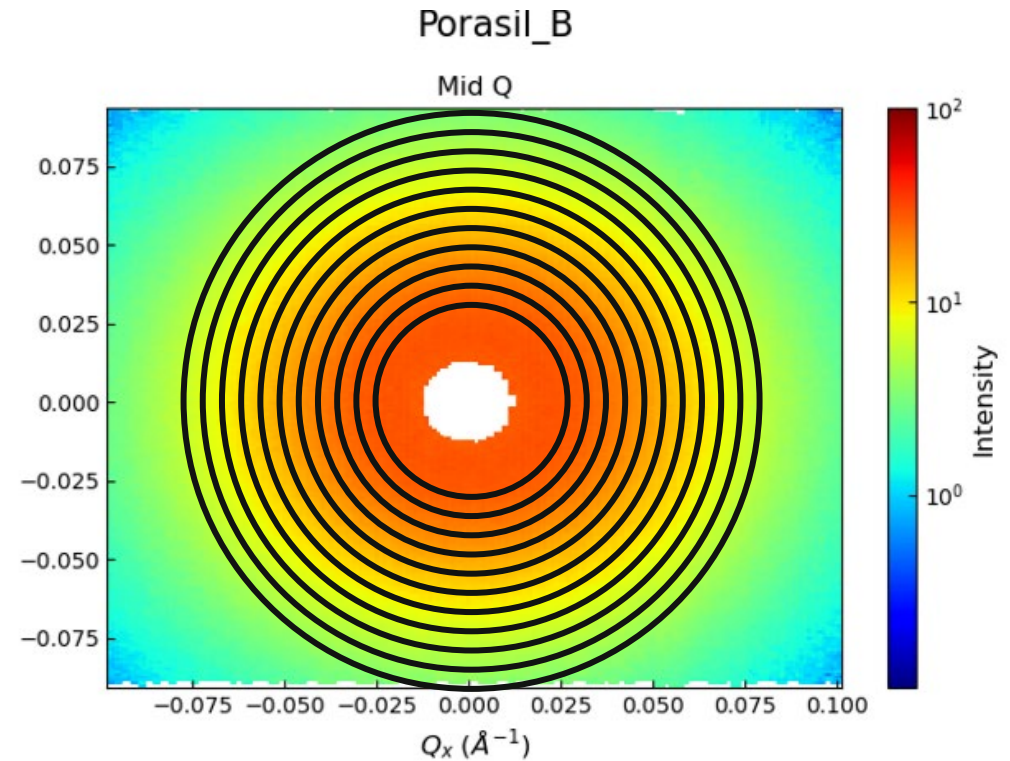


Microstructure

SANS results: Unimer State

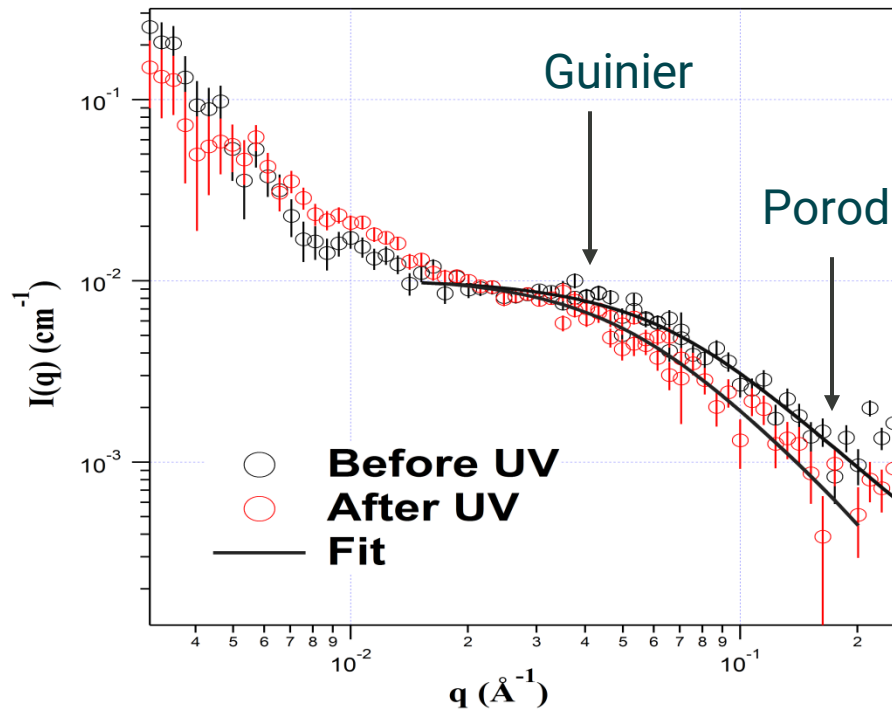


- Guinier Approximation at low- q :
 - $I(Q) = I(0) \exp(-Q^2 R_g^2/3)$
 - $\ln[I(Q)]$.vs. Q^2 plot where $Q.R_g < 1.0$
 - $R_g = \sqrt{3 \cdot \text{slope}}$
 - $M = (1000 \cdot I(0) \cdot d^2 \cdot N_A) / (C \cdot \Delta \rho^2)$
- Power-law at high q



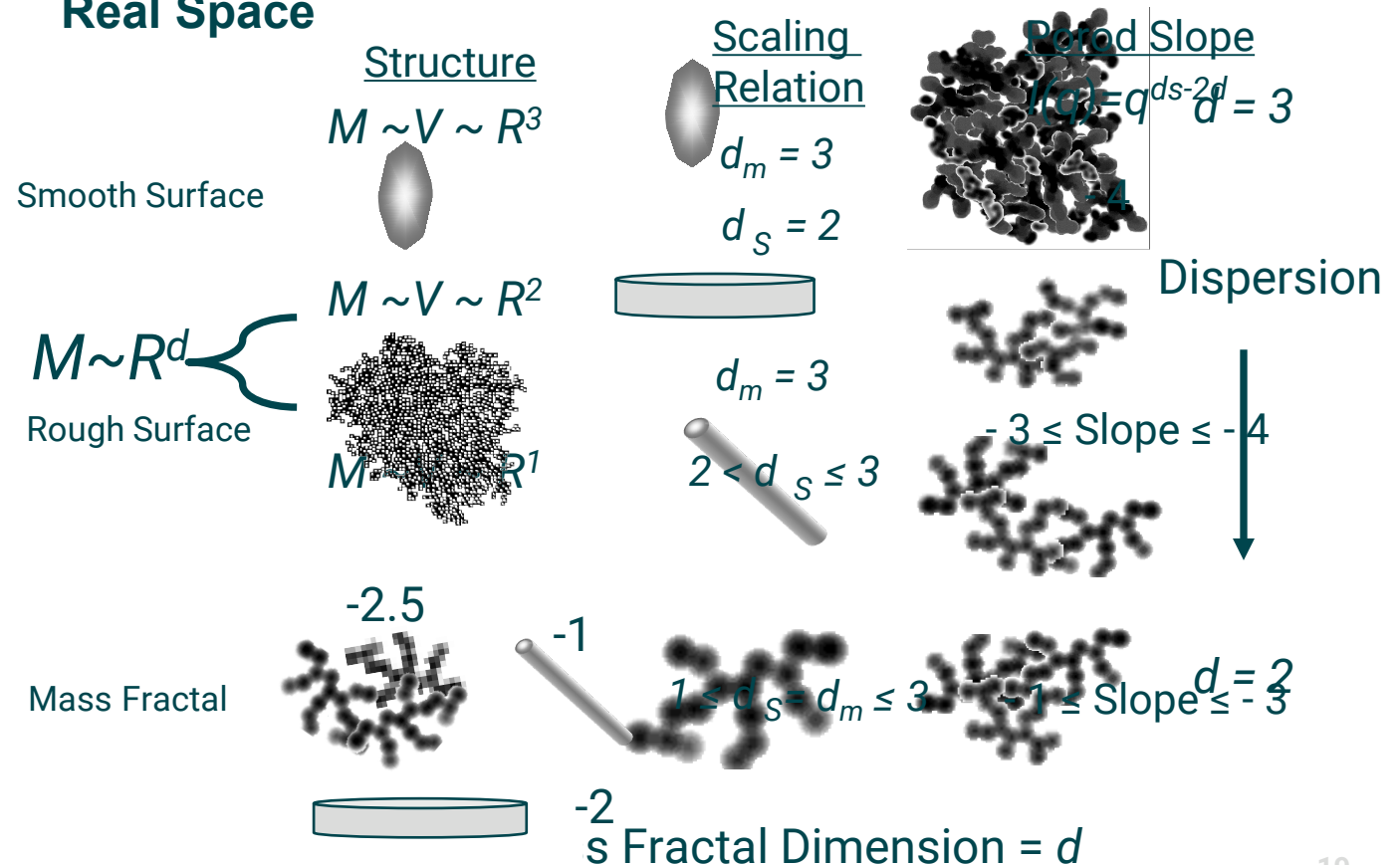
Lilin He, et al. **Polymer**, Volume 105, 25-34 (November 2016) <https://doi.org/10.1016/j.polymer.2016.10.019>

SANS results: Unimer State



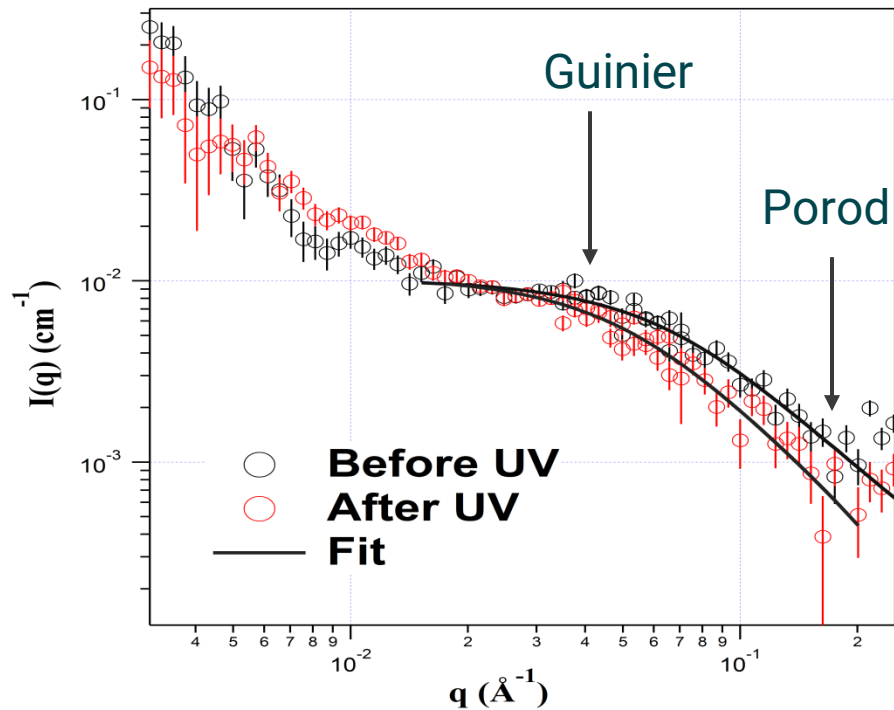
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 - $R_g = \sqrt{(3 \cdot \text{slope})}$
 - $M = (1000 \cdot I(0) \cdot d^2 \cdot N_A) / (C \cdot \Delta \rho^2)$
- Power-law at high q

Real Space



Lilin He, et al. **Polymer**, Volume 105, 25-34 (November 2016) <https://doi.org/10.1016/j.polymer.2016.10.019>

SANS results: Unimer State

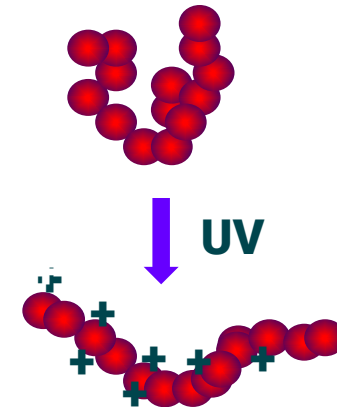


- **0.02wt%**
- **15 °C**

A model for polymers with excluded volume fraction yields R_g and Porod exponent

Before UV
 $R_g = 25.5 \text{\AA}$
Porod exponent = 1.94

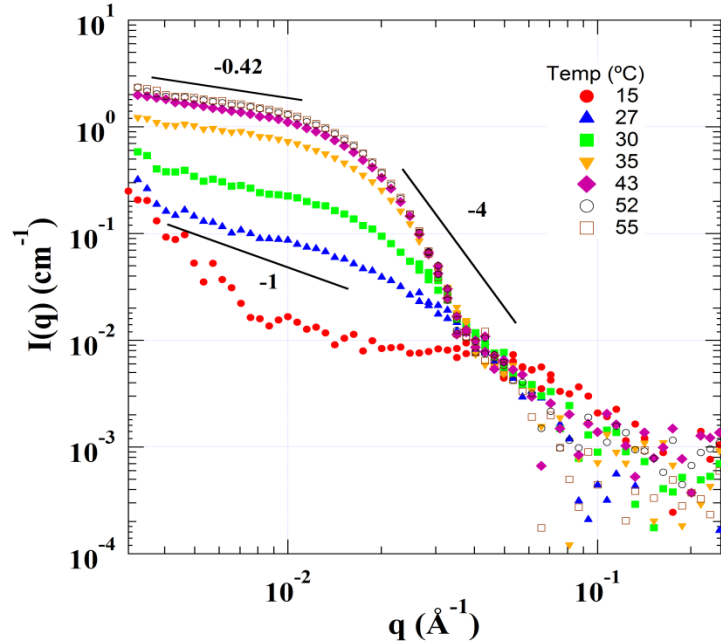
After UV
 $R_g = 31.1 \text{\AA}$
Porod exponent = 1.59



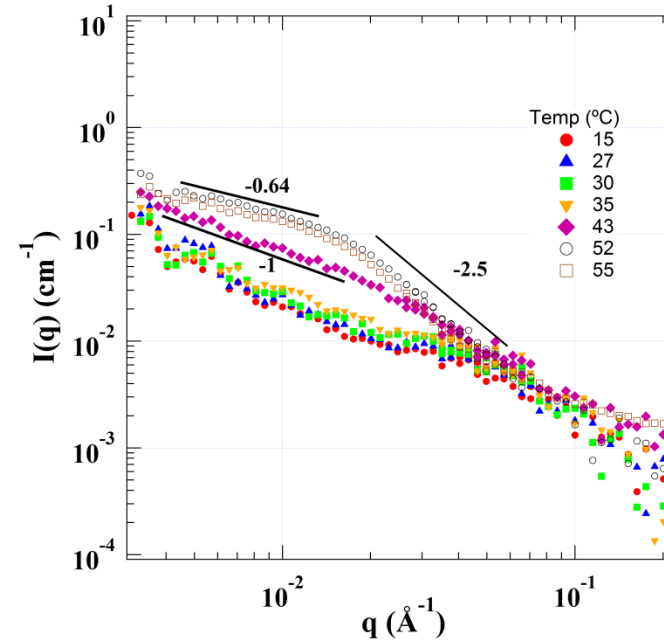
- ***Both solutions contain single chains and loosely-assembled clusters***
- ***The chains are more stretched after UV irradiation***

Temperature Effect: dilute solution

Before UV



After UV



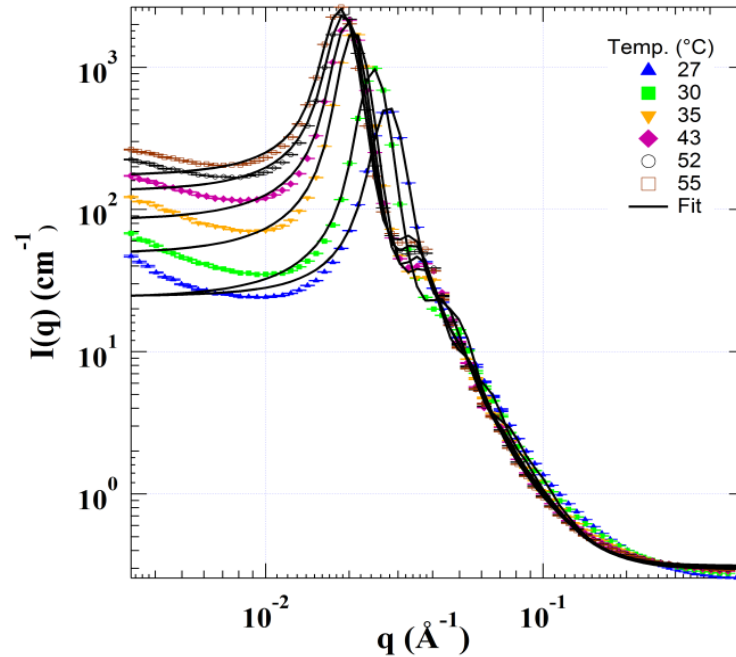
0.02wt%



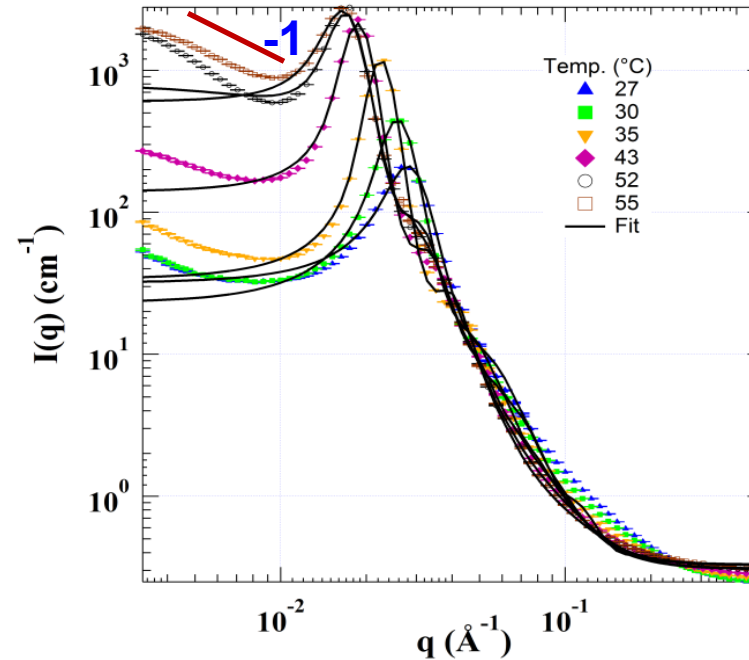
- Micelles form above critical micelle temperature
- Cleaving the light sensitive group defers the formation of the micelles

Temperature Effect: Concentrated solution

Before UV



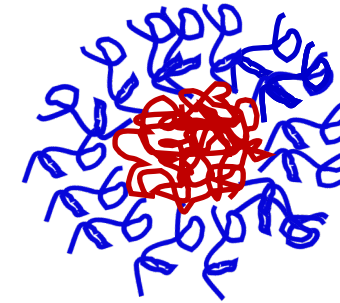
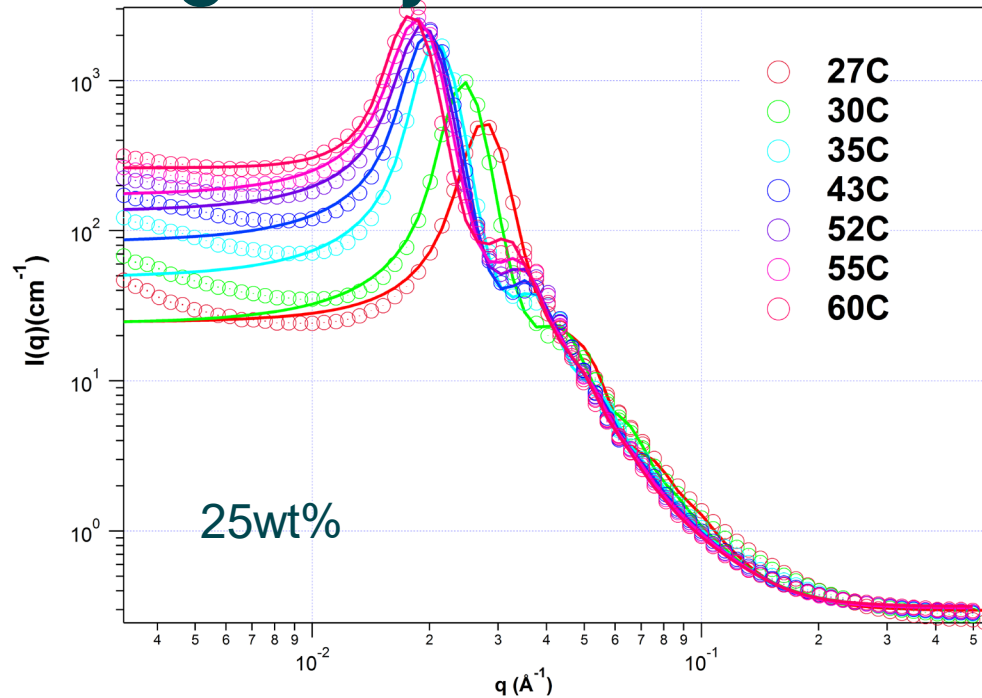
After UV



25 wt%

- Micelles grows with temperature;
- Possible sphere to rod transition at 55C for the UV exposed sample.

Model Fitting: Polycoreshell model



$$\frac{d\Sigma(Q)}{d\Omega} = \Delta\rho^2 \phi V_p P(Q) S(Q)$$

Percus-Yevick closure of the Ornstein-Zernike equation

Material Balance Calculation

$$N_{ag}(131 \cdot v_{TN} + 113 \cdot v_{PEO} \cdot f) + 30 \cdot c = \frac{4\pi}{3} R_A^3$$

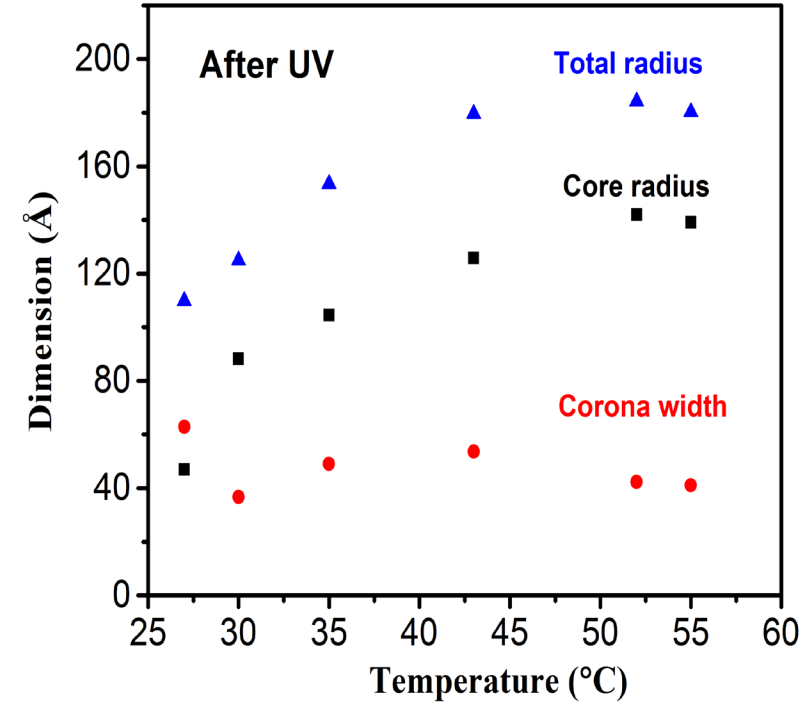
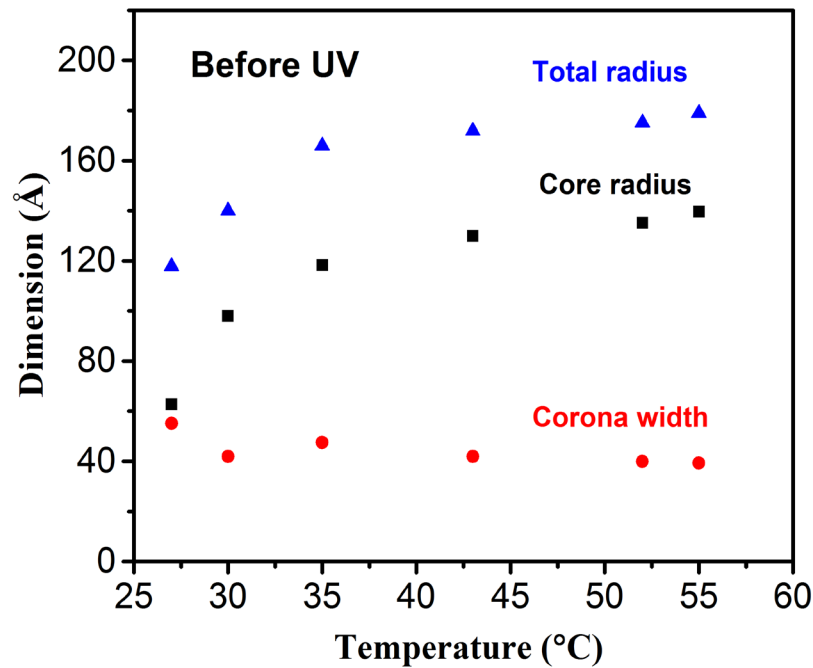
$$N_{ag} \cdot 113 \cdot v_{PEO} \cdot (1-f) + 30 \cdot d = \frac{4\pi}{3} (R_B^3 - R_A^3)$$

$$N_{ag}(131 \cdot b_{TN} + 113 \cdot b_{PEO} \cdot f) + 191 \cdot c = \frac{4\pi}{3} R_A^3 \cdot \rho_A$$

$$N_{ag} \cdot 113 \cdot b_{PEO} \cdot (1-f) + 191 \cdot d = \frac{4\pi}{3} (R_B^3 - R_A^3) \cdot \rho_B$$

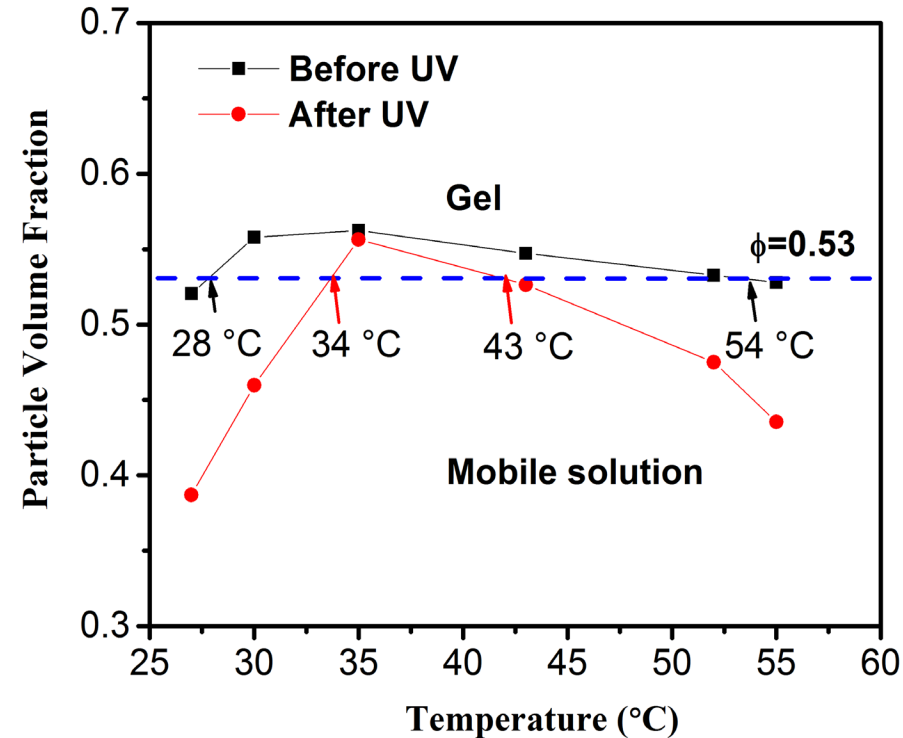
- **Micelle size (core and corona) and volume fraction**
- **Size polydispersity**
- **SLDs in core and corona**
- **Aggregation number**
- **Number of D₂O molecules in core and corona**
- **Fraction of PEO chains in core and corona**

Temperature Effect: Concentration solution



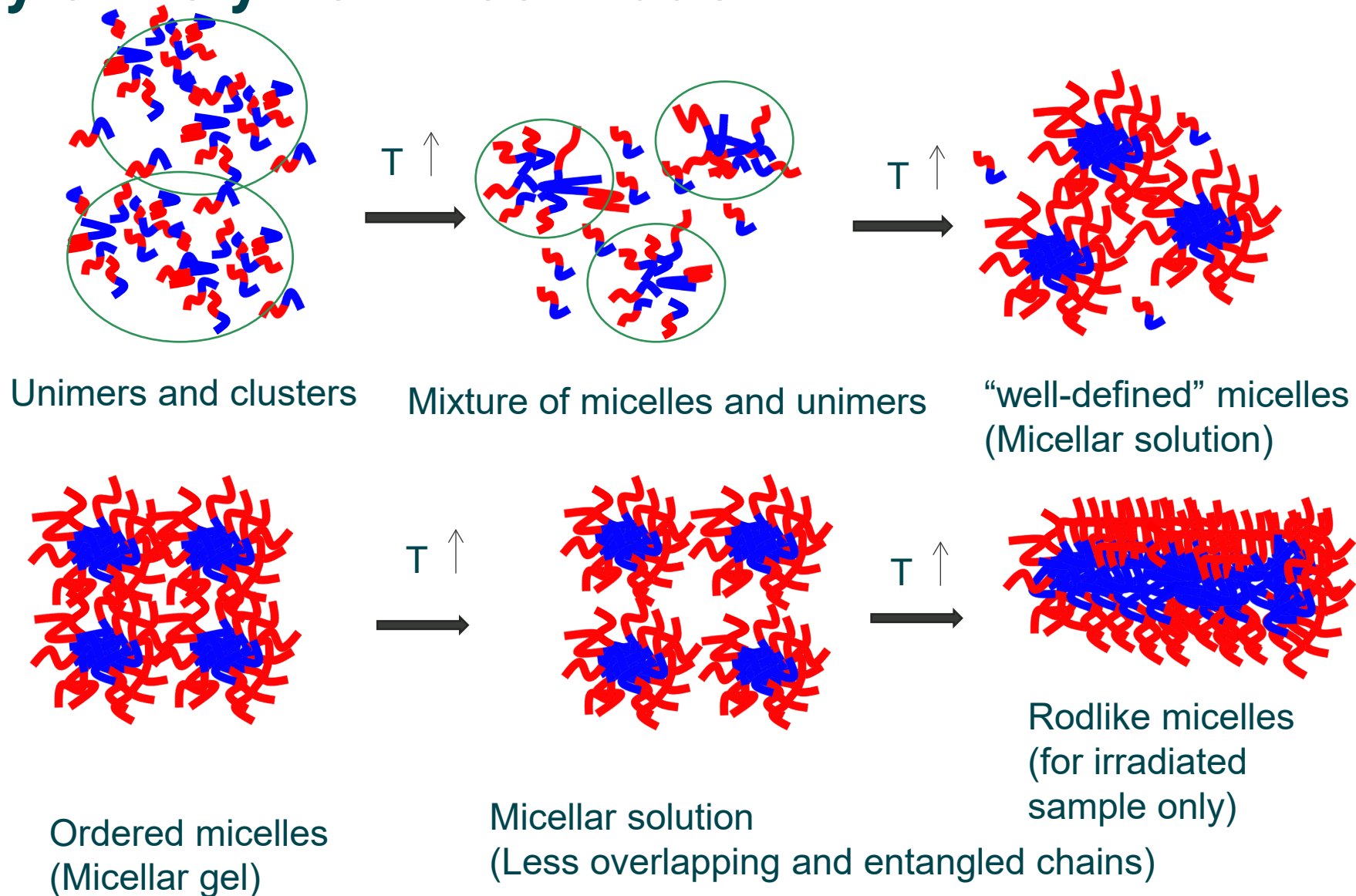
- The core size increases while the corona width remains;
- The slight shrinkage of corona at high temperatures is attributed to the loss of water, which is caused by the reduced miscibility of water and PEO.

Temperature dependence of particle volume fraction



- Micelle effective volume fraction agrees the viscoelastic properties of solutions;
- Delayed gel formation in irradiated sample due to higher LCST transition;
- Critical value 0.53 ± 0.02

Summary of Polymer Micellization



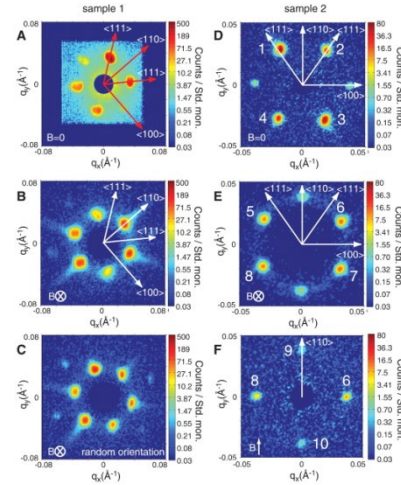
Realization of magnetic skyrmions in thin films at ambient conditions

Ryan Desautels, Lisa DeBeer-Schmitt, Sergio Montoya, Julie Borchers, Soong-Guen Je, Nan Tang, Mi-Young Im, Micheal Fitzsimmons, Eric Fullerton, Dustin Gilbert

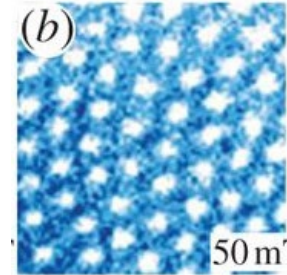
Skyrmions: a brief introduction



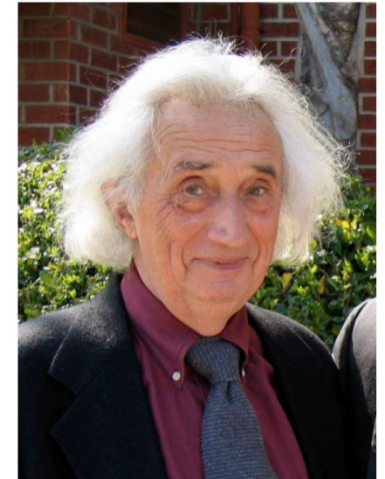
Tony Skyrme
Nuc. Phys. 13, 556 (1962)



Science 323, 915 (2009)

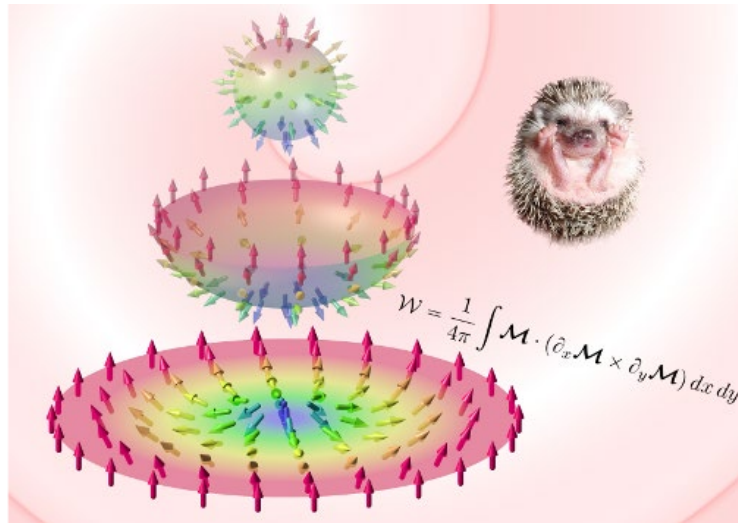


Nature 465, 901 (2010)

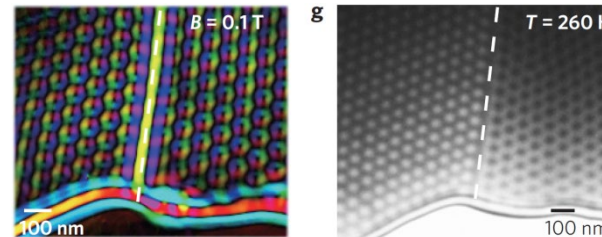


Igor Dzyaloshinskii
Soviet Physocs Jetp. 5, 1259 (1957)

Toru Moriya
Phys. Rev. 120, 91 (1960)



Predicted in magnetic systems
(Phys. Rev. Lett. 87, 037203 (2001))



Nature Mater. 10, 106 (2011)

Direct Exchange Prefers
Parallel Alignment

$$E \propto J(\vec{S}_1 \cdot \vec{S}_2)$$

$$E \propto D(\vec{S}_1 \times \vec{S}_2)$$

DM Interaction Prefers
90° Orientation

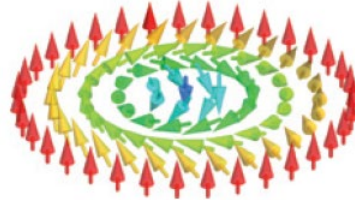
Defines a 'handedness'

Skyrmions: a brief introduction

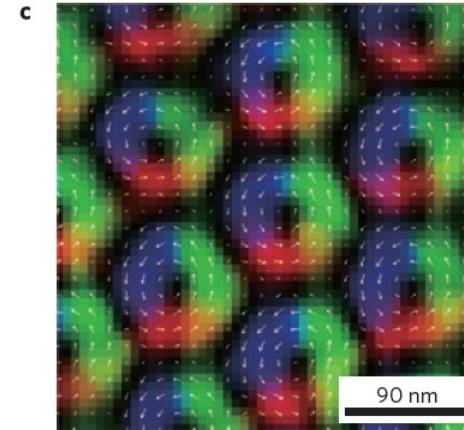
Néel (hedgehog)



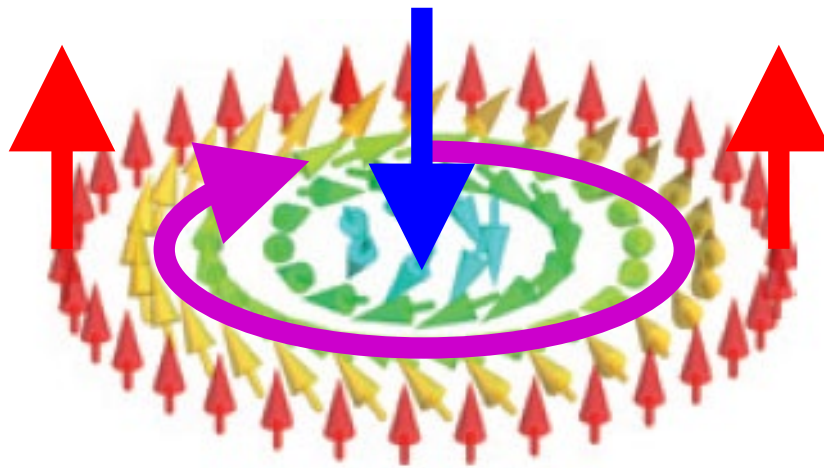
Bloch



(arrows indicate magnetic spins)



Fert, Cros, Sampaio, Nat. Nanotechnol. **8**, 152 (2013).



Circularity (CW and CCW)

Polarity (Core-up, Core-down)
anti-parallel to perimeter

Genus 0



Marble

Genus 1



Doughnut

Genus 2



Kettle

Genus 3 or more



Strainer



Bowling Ball



Coffee Cup



Scissors

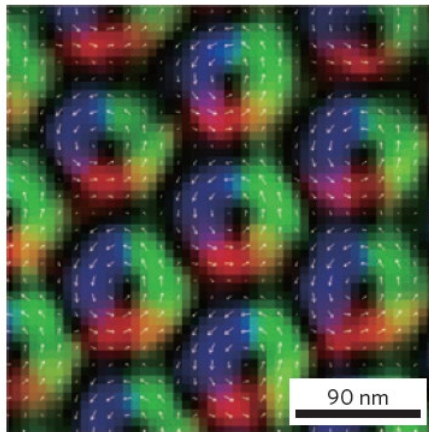
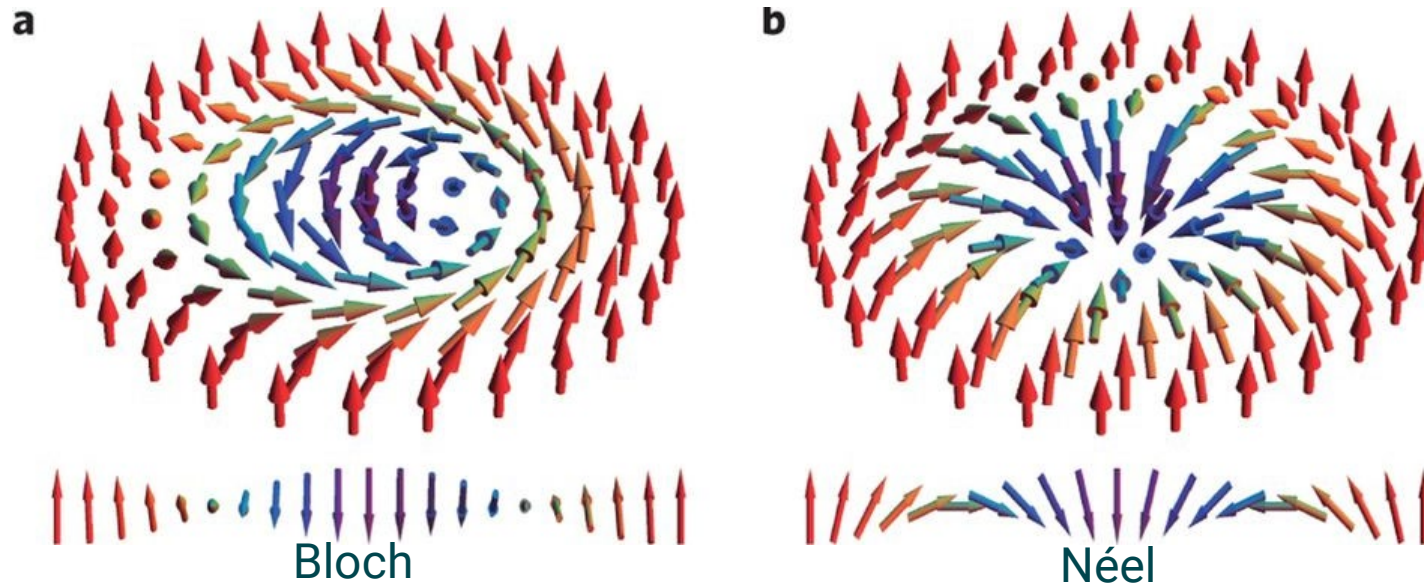


Grater

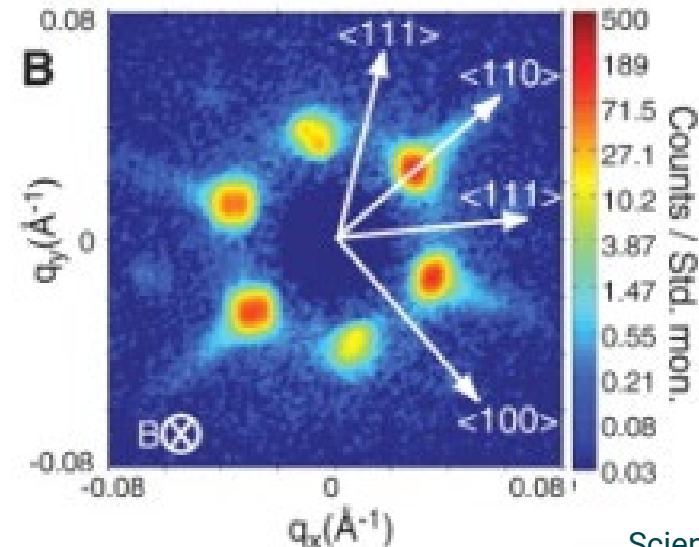
Changing between topological classes
requires irreversible processes

Skyrmions: a brief introduction

I. Kezsmarki et al, *Nature Materials* **volume14**, pages1116–1122 (2015)



Fourier Transform



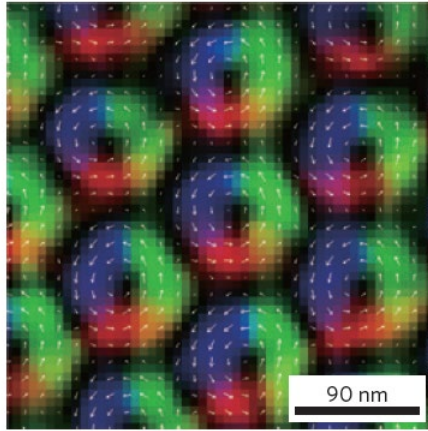
Fert, *Nat. Nanotechnol.* **8**, 152 (2013).

Science 323, 915 (2009)

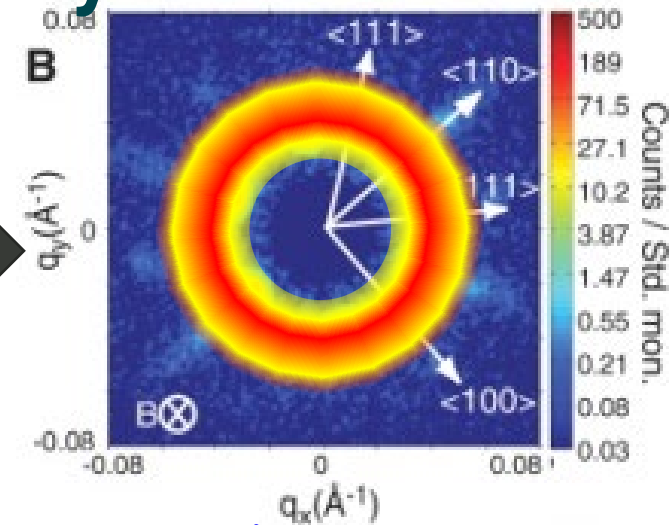
Magnetism effects on Skymrion stability

Science 323, 915 (2009)

Fert, Nat. Nanotechnol. **8**, 152 (2013).



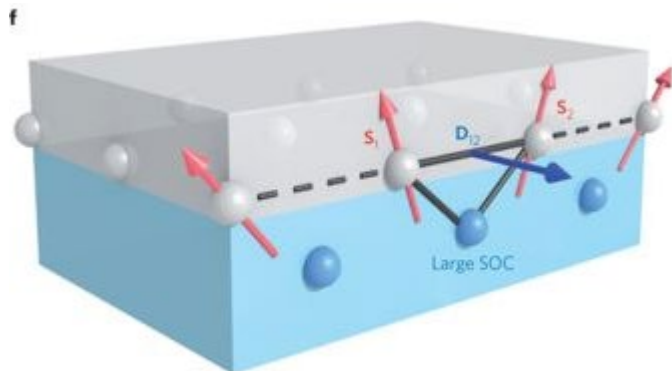
Fourier Transform



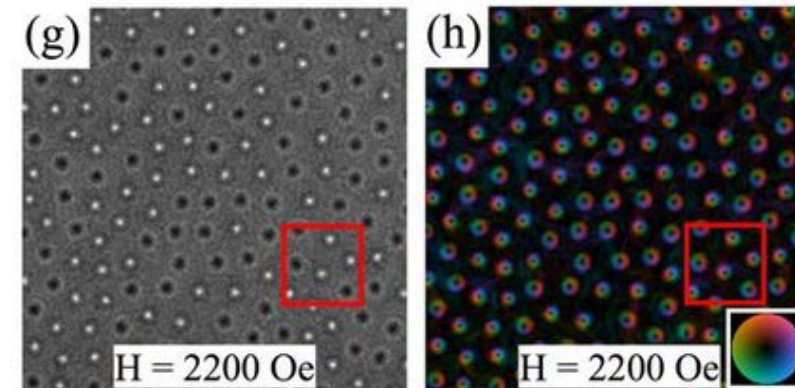
Imagine if we didn't have this long-range ordering...

Origin of the long-range order is in magneto-crystalline coupling

We are interested in skyrmion systems which have weak magnetocrystalline coupling

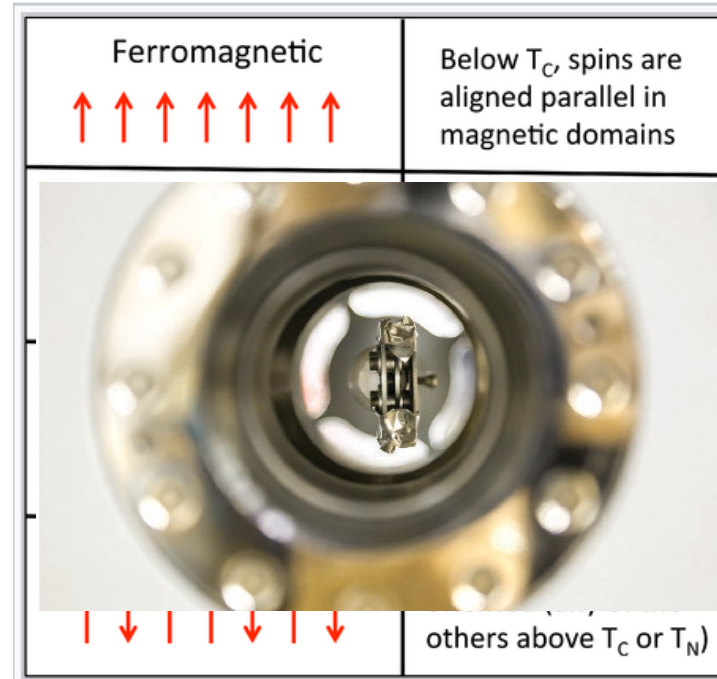
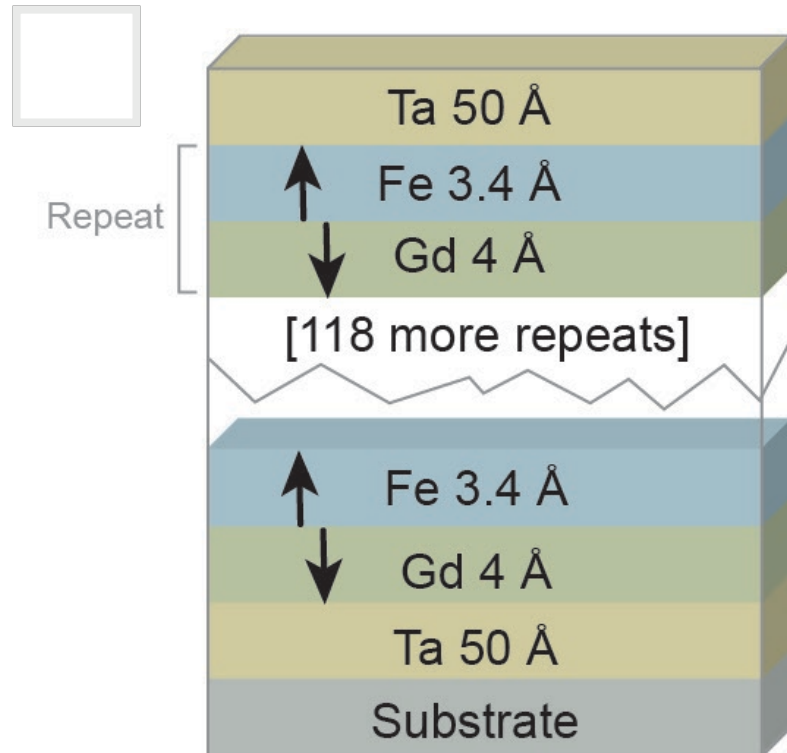


Fert *et al.*, Nat. Nanotechnol. **8**, 152 (2013)
OAK RIDGE
National Laboratory



Montoya *et al.*, Phys. Rev. B **95**, 024415 (2017)

Fe/Gd Multilayer Thin Films: The Ingredients for Skyrmions at Ambient Conditions



chem.libretext.org

**Ferrimagnetic construction
with perpendicular
magnetic anisotropy**

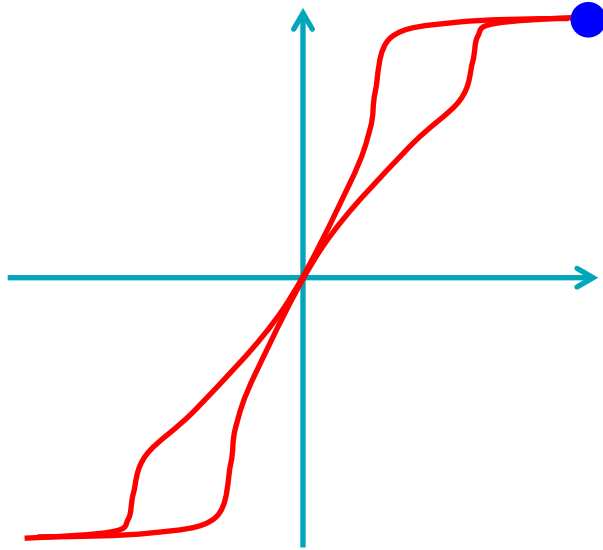
Dipole-stabilized skyrmions

Limited/no DMI

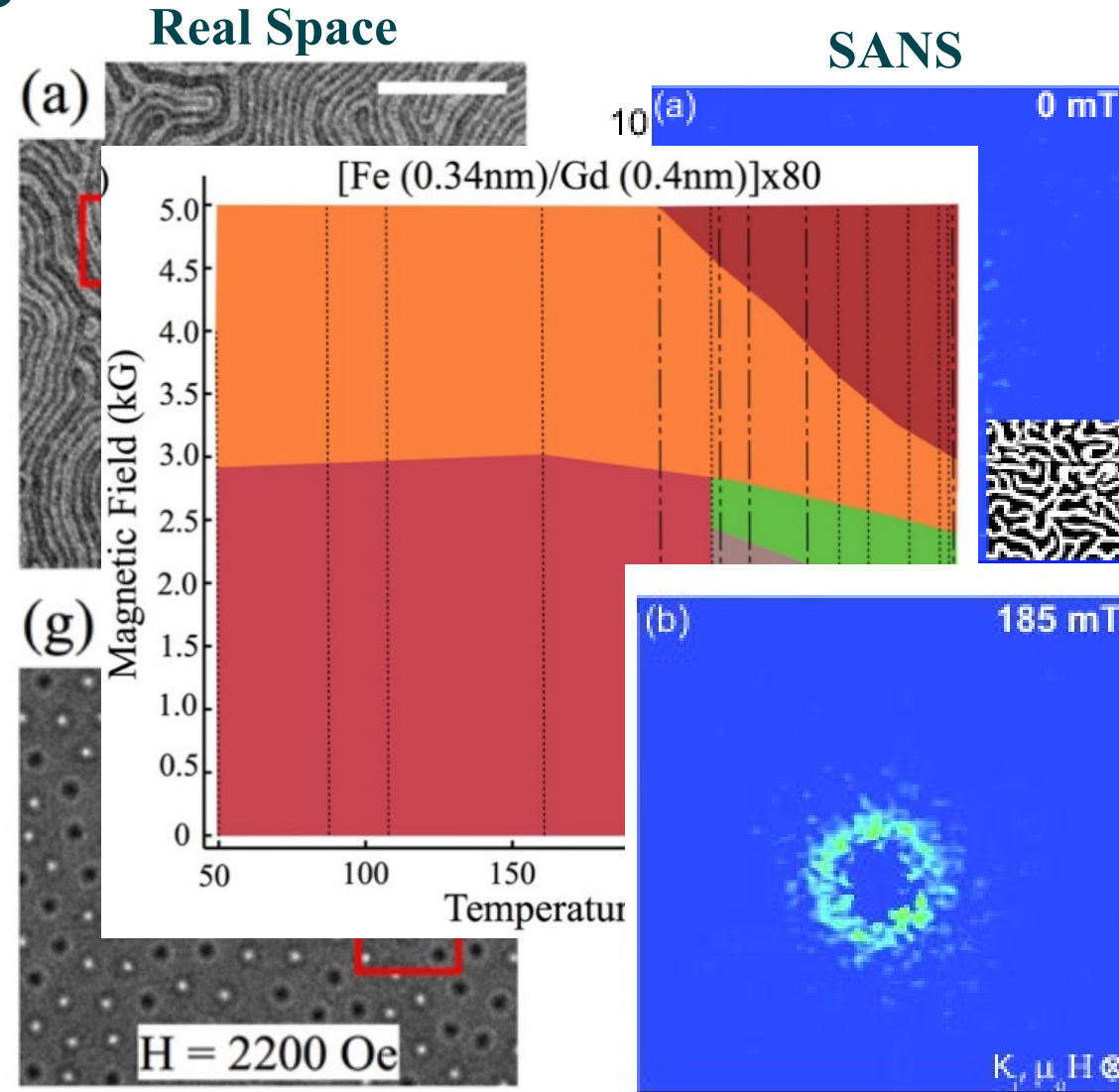
**No in-plane structure to cement
a long-range orientation**

Forming the Skyrmion State

Montoya *et al.*, Phys. Rev. B **95**, 024415 (2017)



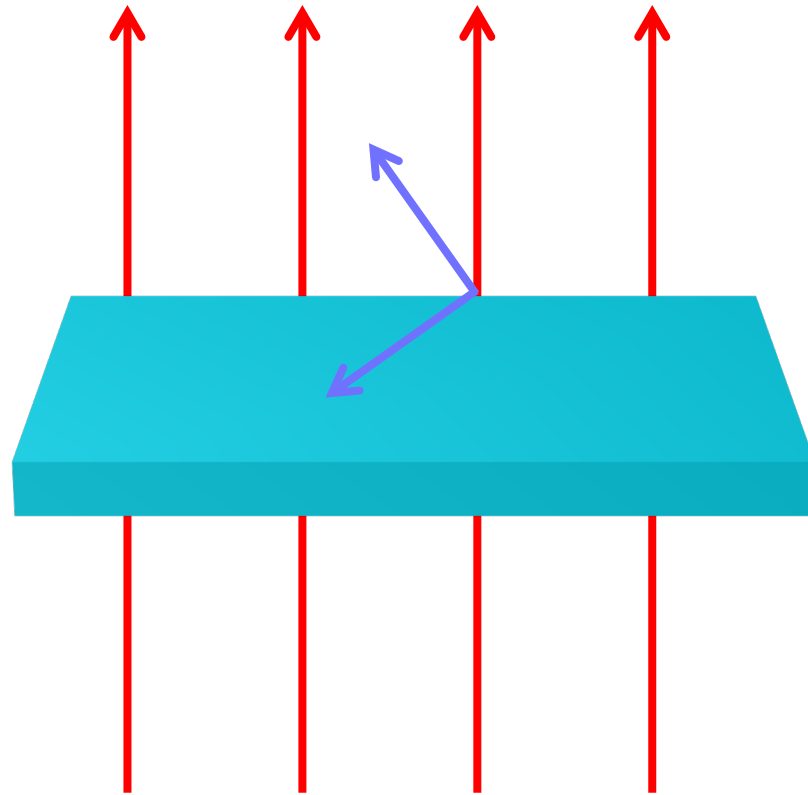
Montoya *et al.*, Phys. Rev. B **95**, 024415 (2017)



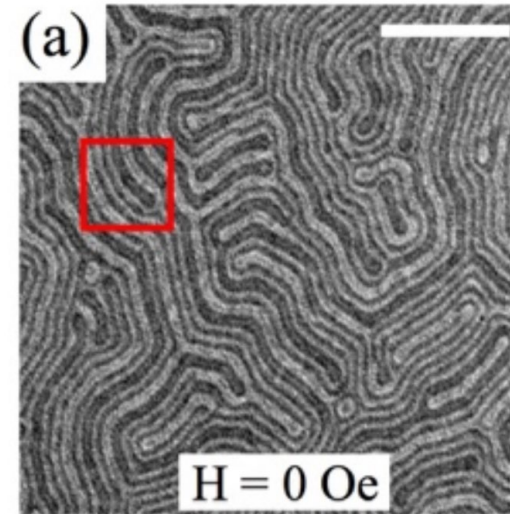
From saturation, worm domain remanent state

Break up into chiral bubbles, no chirality control, no long range order

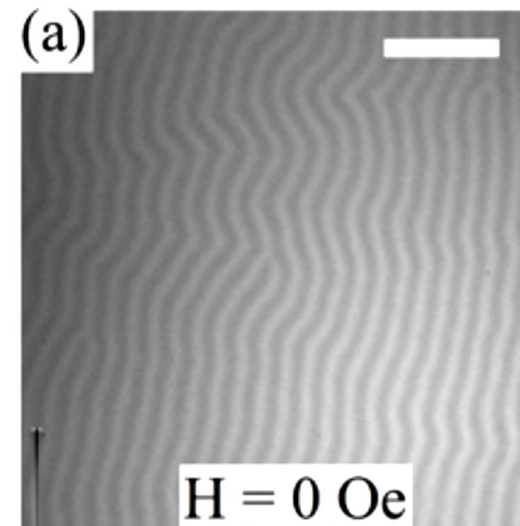
Generating an Artificial Striped Phase



Applied Field



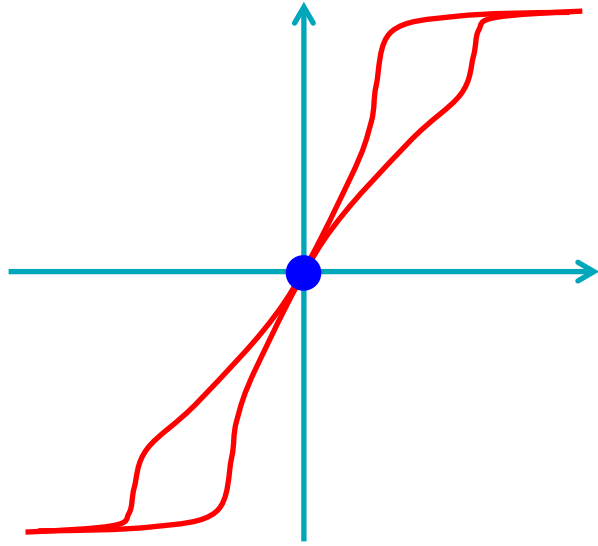
Labyrinth (or worm) domains



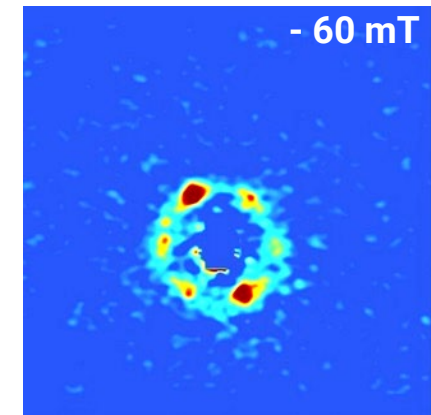
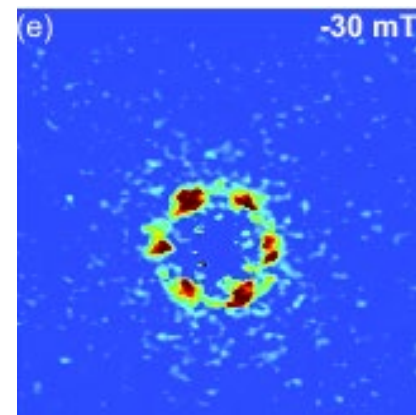
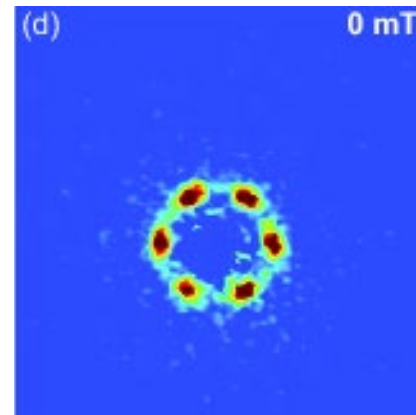
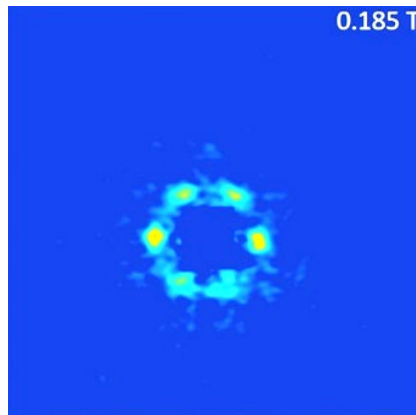
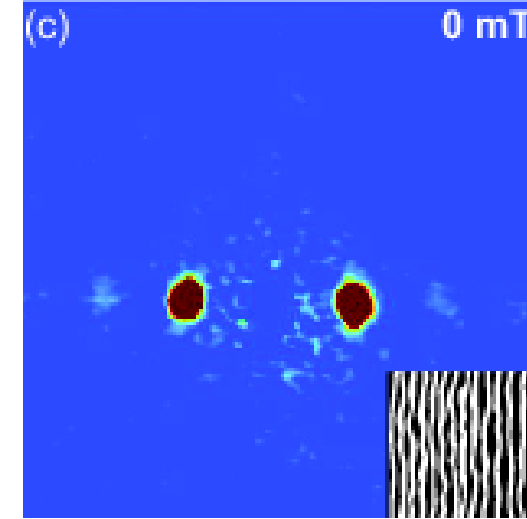
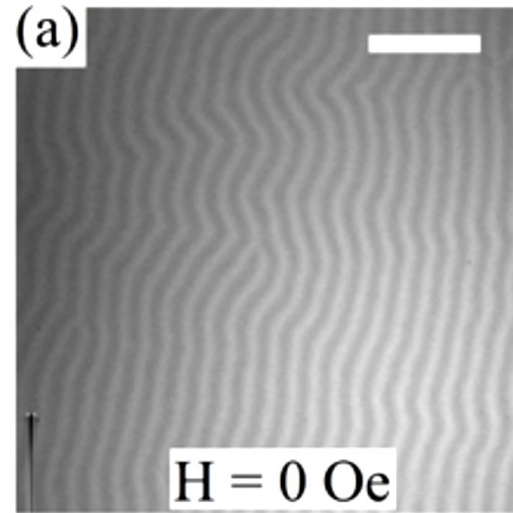
Tilting the sample imparts an in-plane field which breaks the symmetry

Artificial Stripe Domain

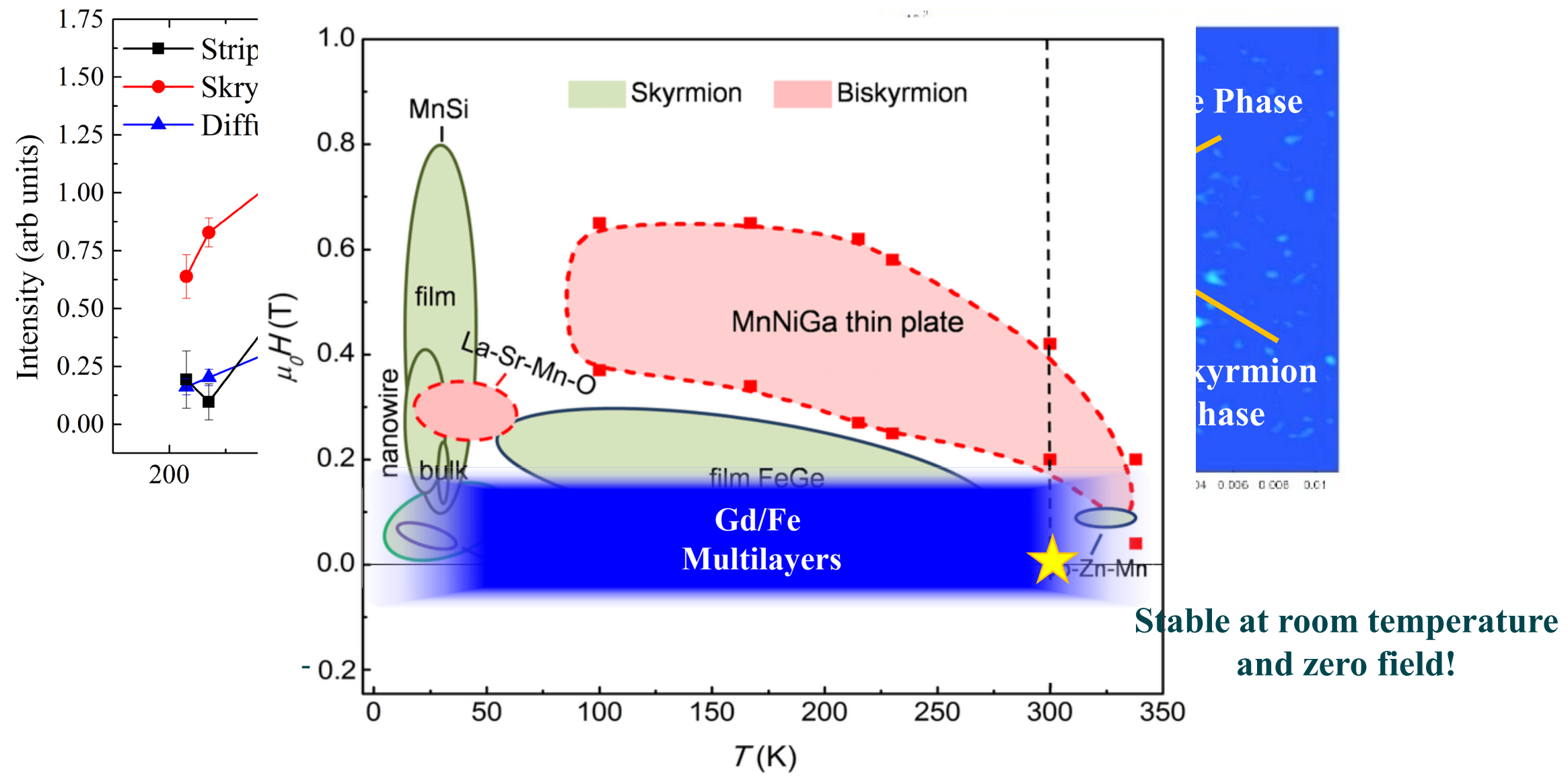
Generating an Artificial Striped Phase



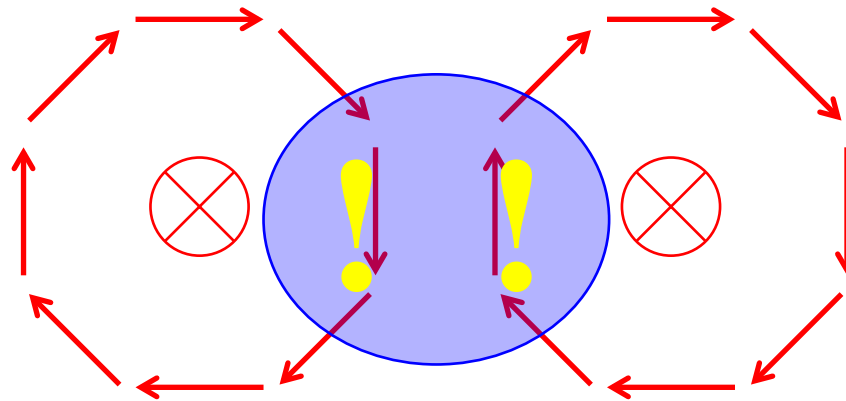
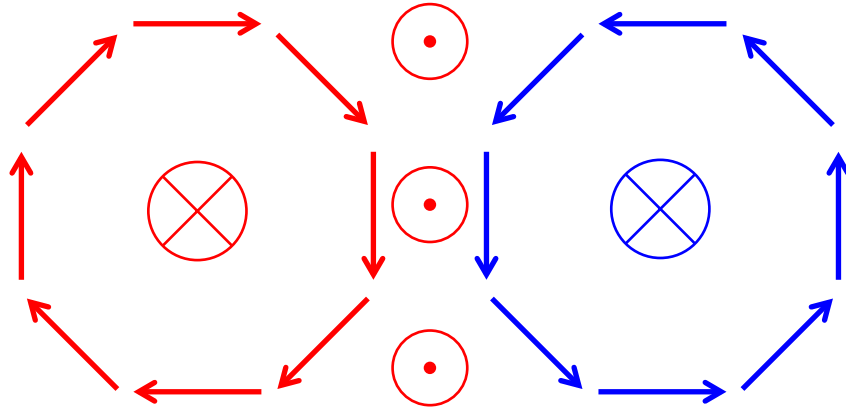
Desautels et al., *Under Review*



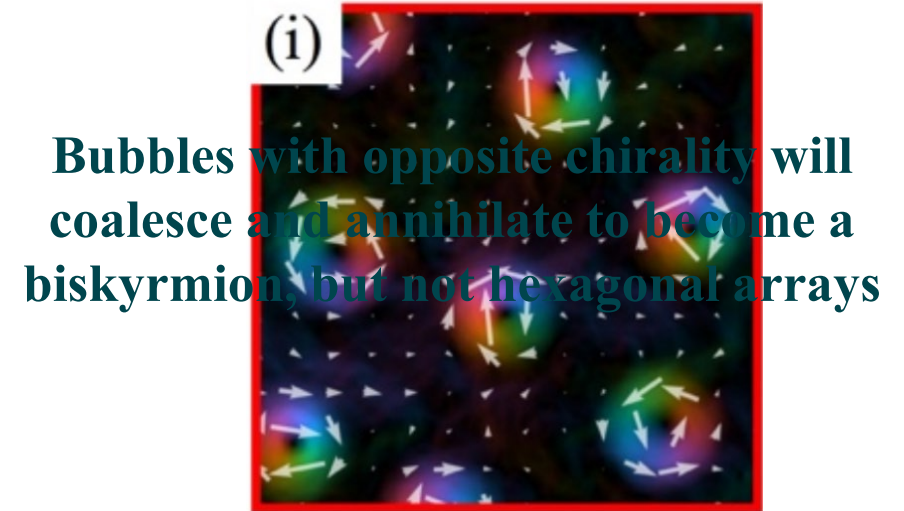
Skyrmion lattice Stability Envelope



Implications of Chirality Control on the stability of Skyrmion Lattices



TEM indicates Bloch-type structure with no circularity control

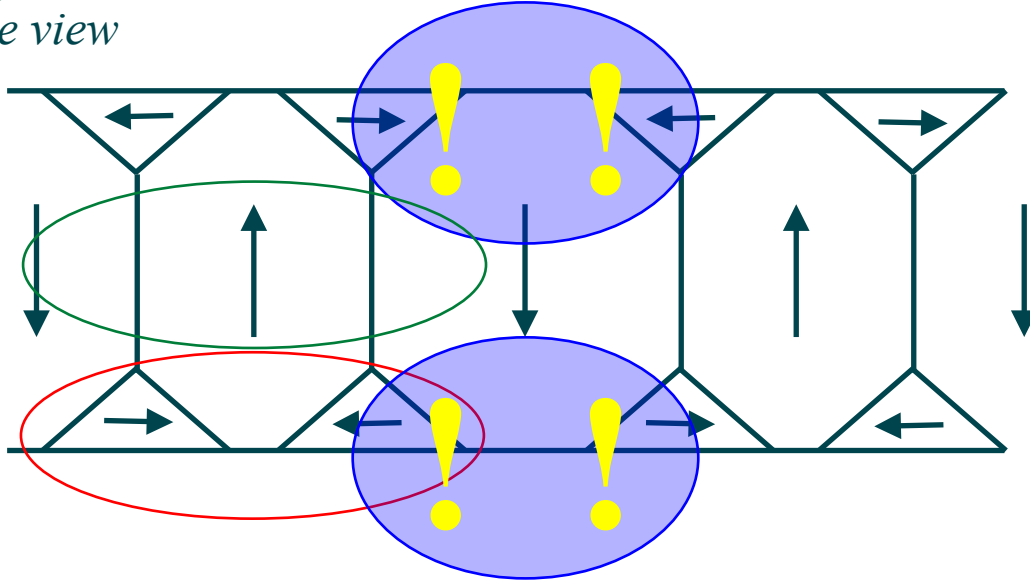


Strong repulsive interactions between bubbles with identical chirality

→ Hexagonally Ordered Arrays

Implications of Chirality Control on the stability of Skyrmion Lattices

Side view



**Appears as Néel Skyrmion, not Bloch
(as observed with TEM)**

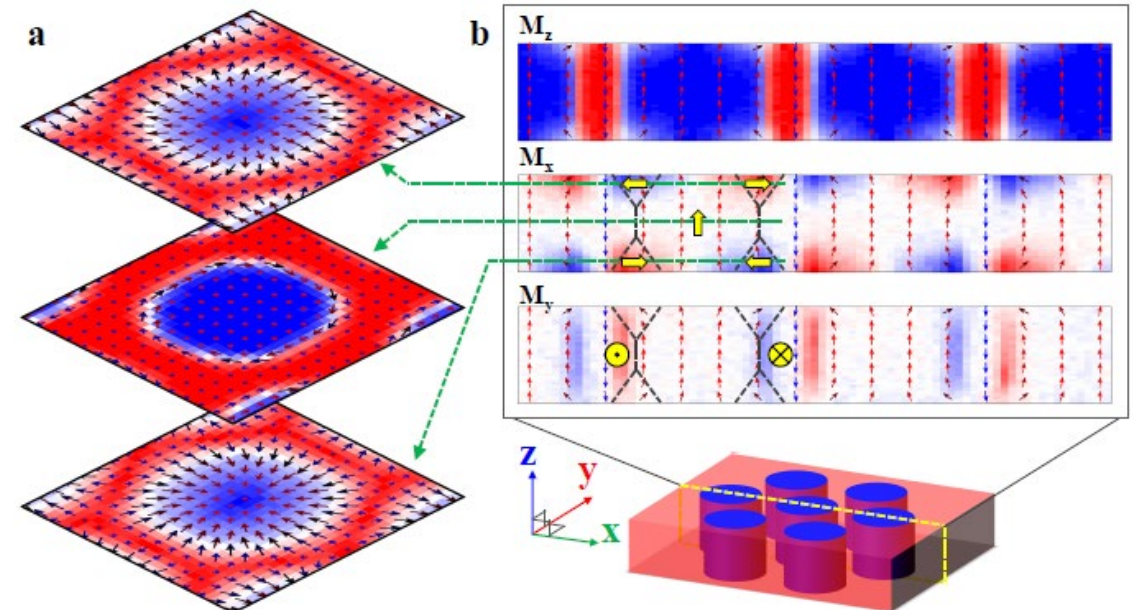
**Maybe Neel on the top and bottom,
Bloch in the middle**

**Without DMI, the Bloch region has no
net chirality (as observed in TEM)**

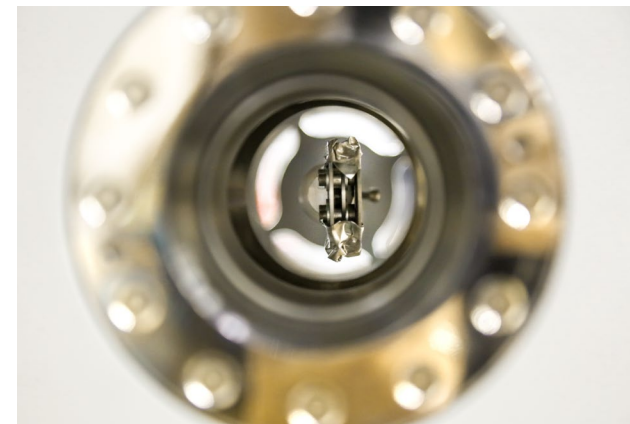
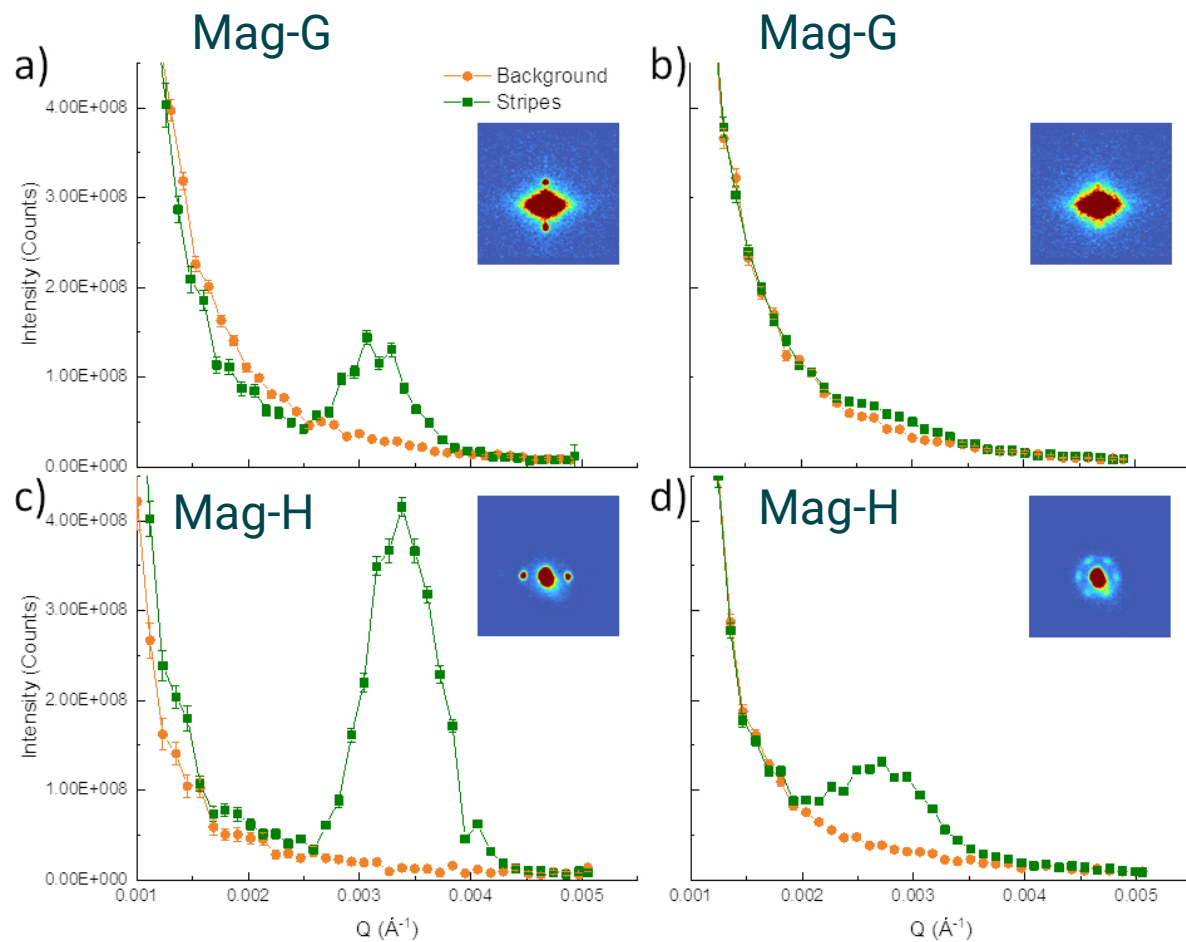
Assumed these are flux-closure
domains with no defined chirality

Strong Repulsive Interaction

Repulsive interactions due to surface features,
which have their chirality defined by the dipole
fields

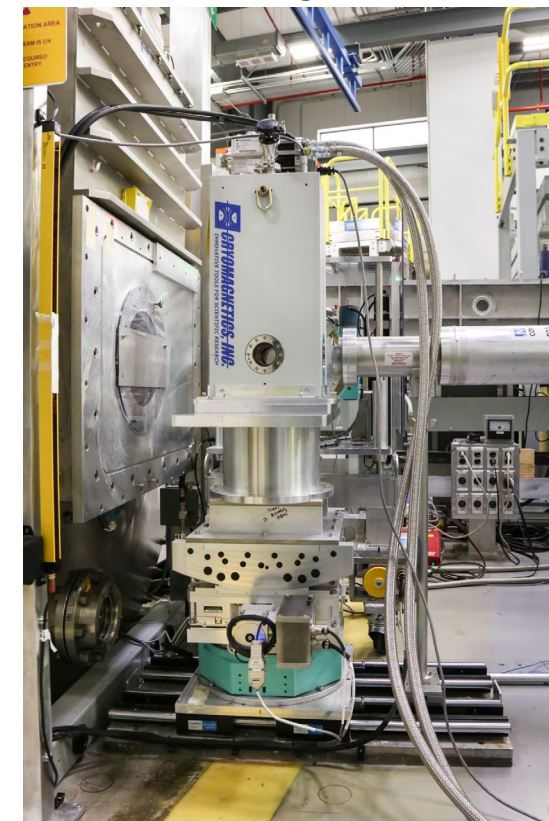
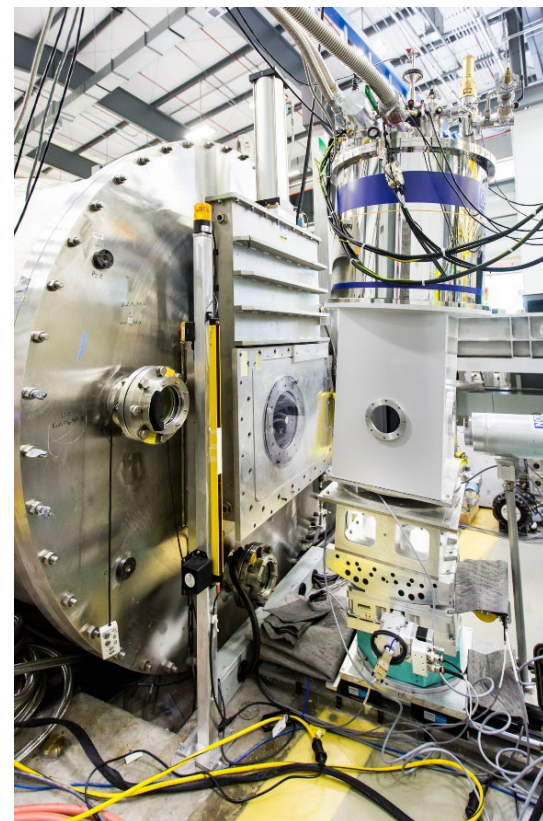


Thin Films studies at GP-SANS



Mag-G

Mag-H



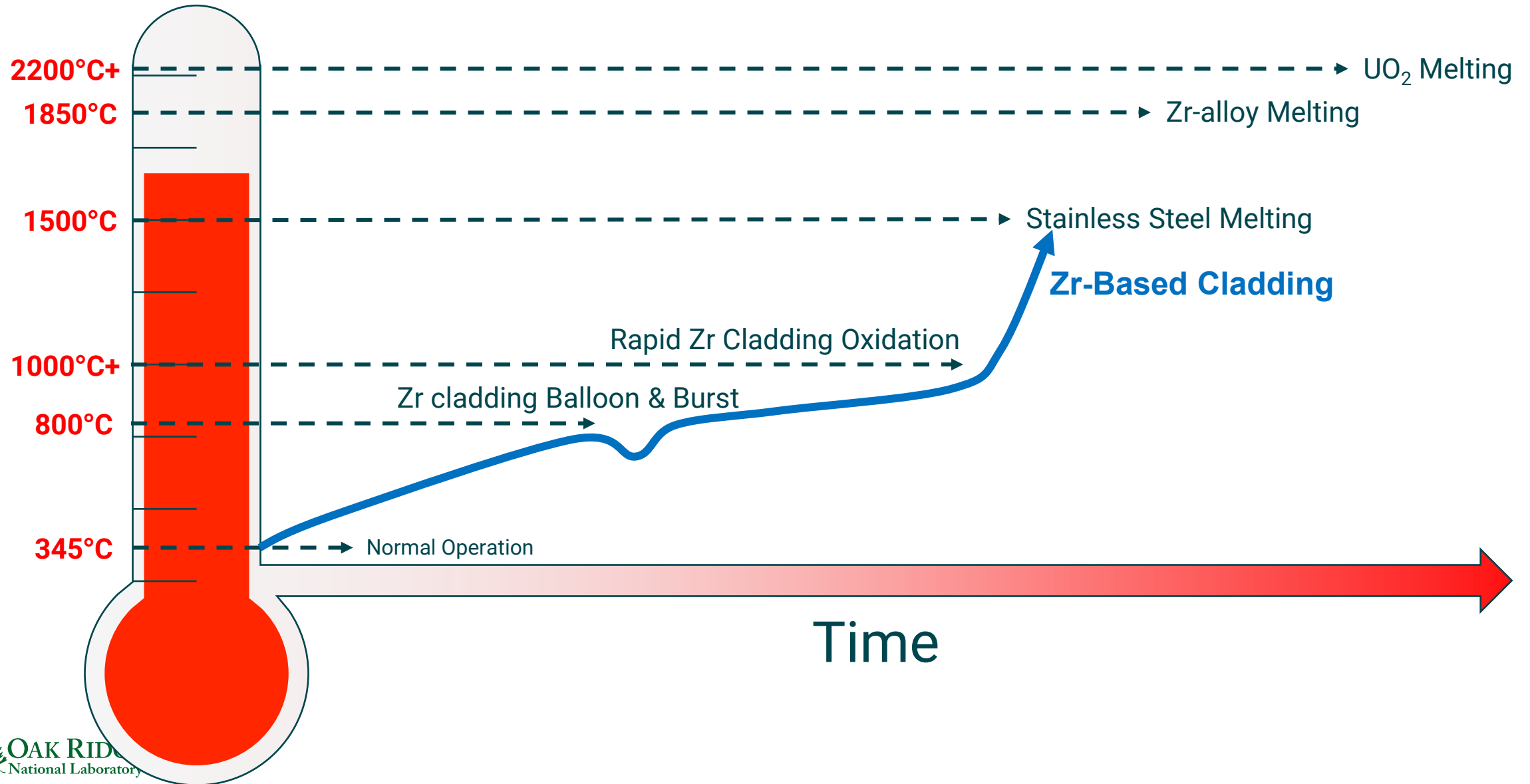
Use of Small-Angle Neutron Scattering to Characterize Novel Steels for Reactor Applications

Kevin G. Field¹, Kenneth C. Littrell^{1*}, and Samuel A. Briggs²

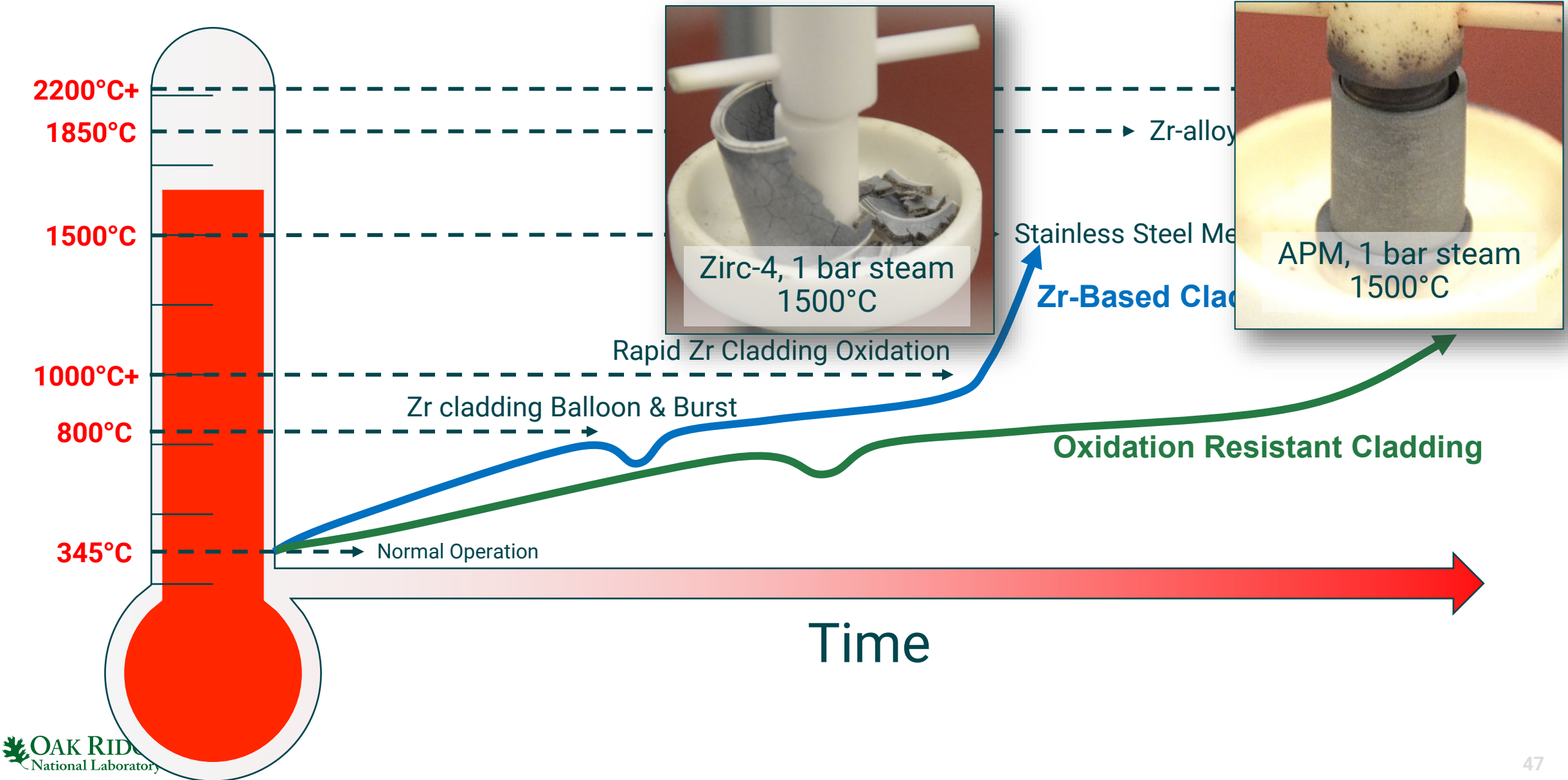
¹*Oak Ridge National Laboratory*

²*Sandia National Laboratories*

Oxidation of cladding is key towards core degradation during a coolant-limited accident scenario



Oxidation of cladding is key towards core degradation during a coolant-limited accident scenario



SANS Scattered Intensity: Local Monodispersed Approximation¹

$$\frac{d\sigma}{d\Omega}(q) = \Delta\rho^2 \int_0^\infty \phi(q, R)^2 S[q, R_{HS}(R)] N(R) dR + Aq^{-B} + C$$

- Contrast: $\Delta\rho^2 = (\rho_{particle} - \rho_{matrix})^2$
- Form factor (for spheres):

$$\phi(q, R) = 3V_o[\sin(qR) - qR\cos(qR)]/(qR)^3$$

- Structure factor: $S[q, R_{HS}(R)] = [1 + 24\eta_{HS}G(R_{HS}q)/(R_{HS}q)]^{-1}$
- Size distribution (Weibull density function):

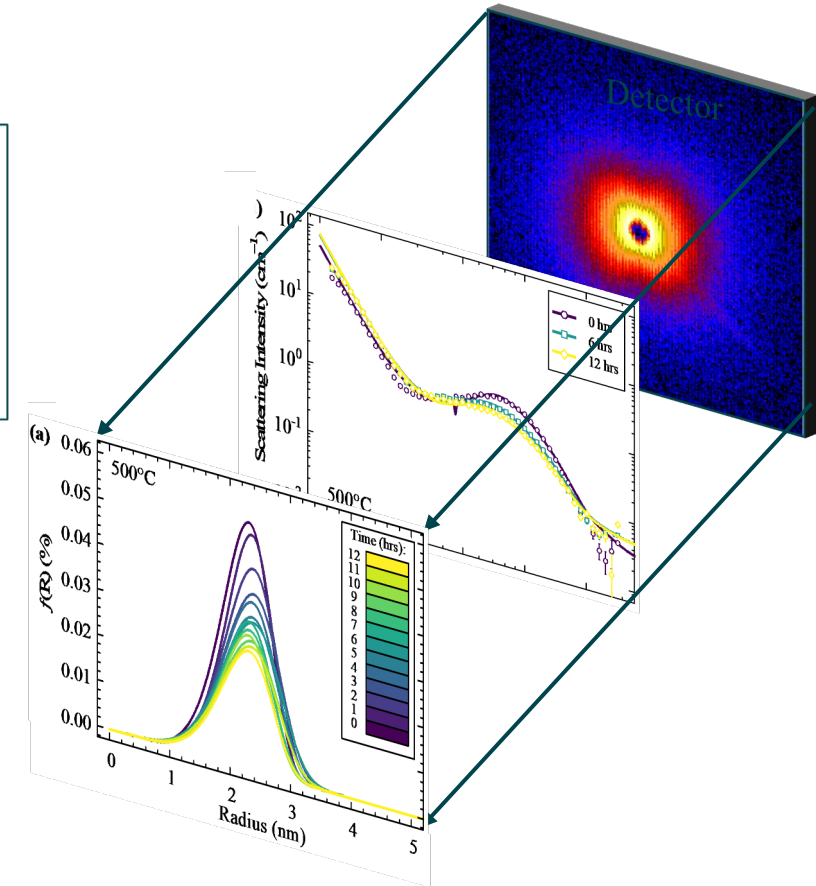
$$N(R) = (R/\bar{R})^{b-1} \exp[-(R/\bar{R})^b]$$

- Low-q power-law:

$$Aq^{-B}$$

- Background:

C



Magnetic Shielded-SANS measurements : Contrast from Neutrons

1. Nuclear contrast:

$$\Delta\rho_{nucl}^2 = (\rho_{nucl,particle} - \rho_{nucl,matrix})^2$$

2. Magnetic contrast:

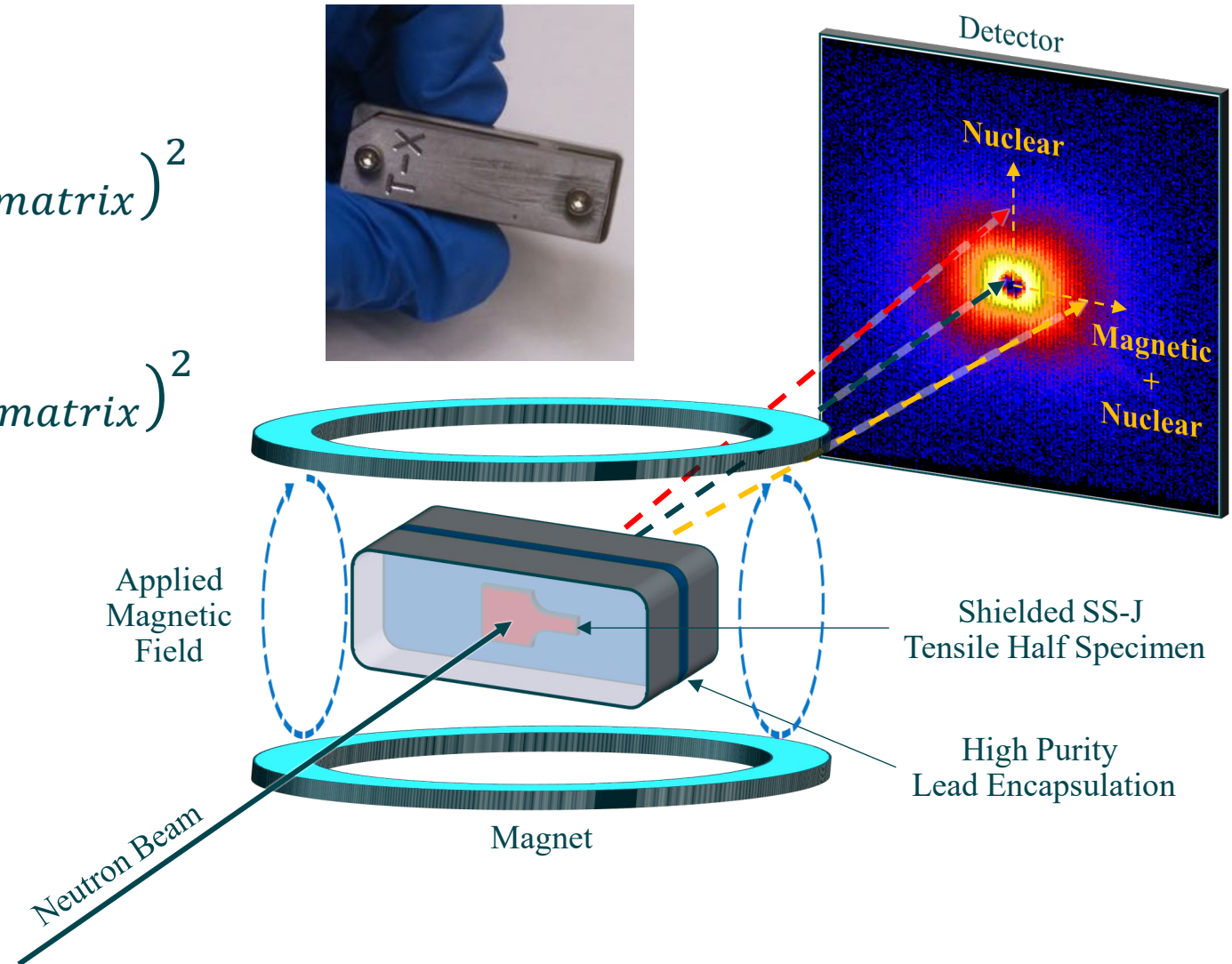
$$\Delta\rho_{mag}^2 = (\rho_{mag,particle} - \rho_{mag,matrix})^2$$

$$\frac{d\sigma}{d\Omega}(q) = \Delta\rho^2 \int_0^\infty \phi(q, R)^2 ... \square$$

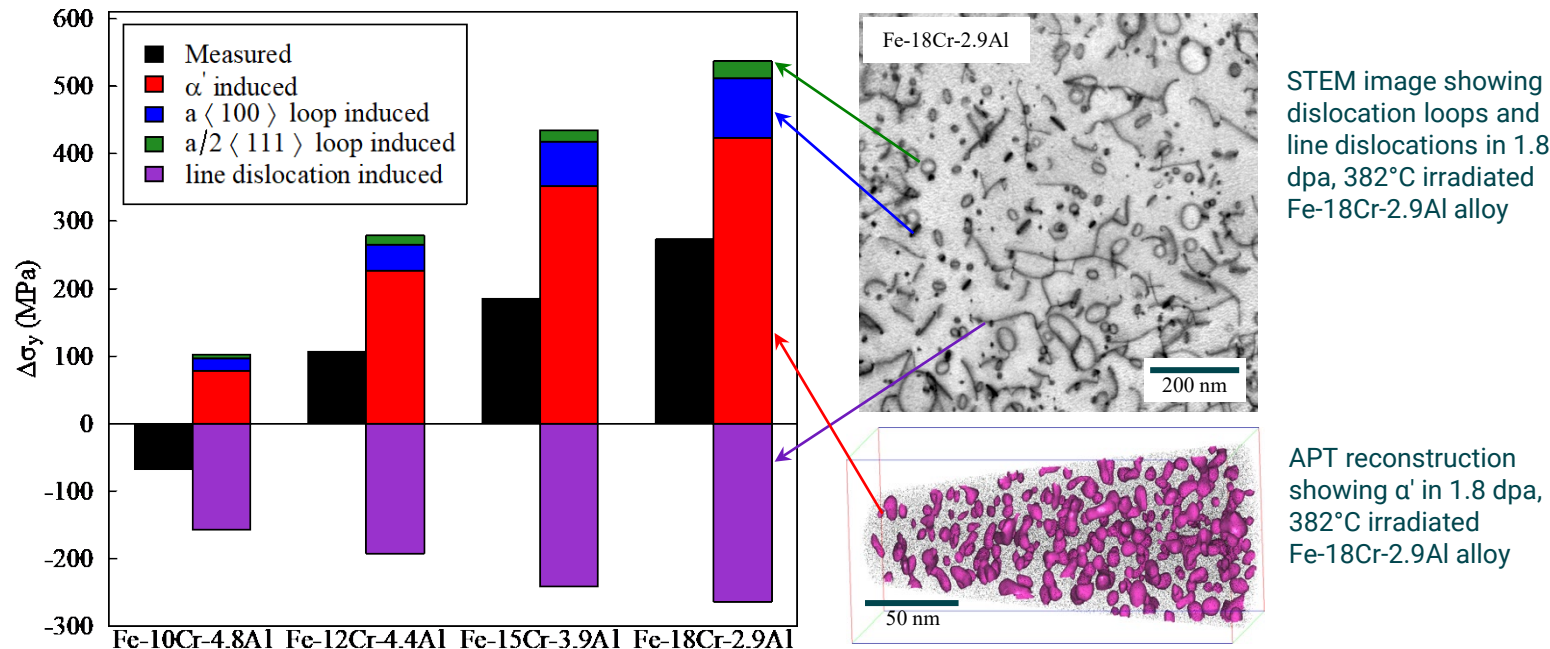
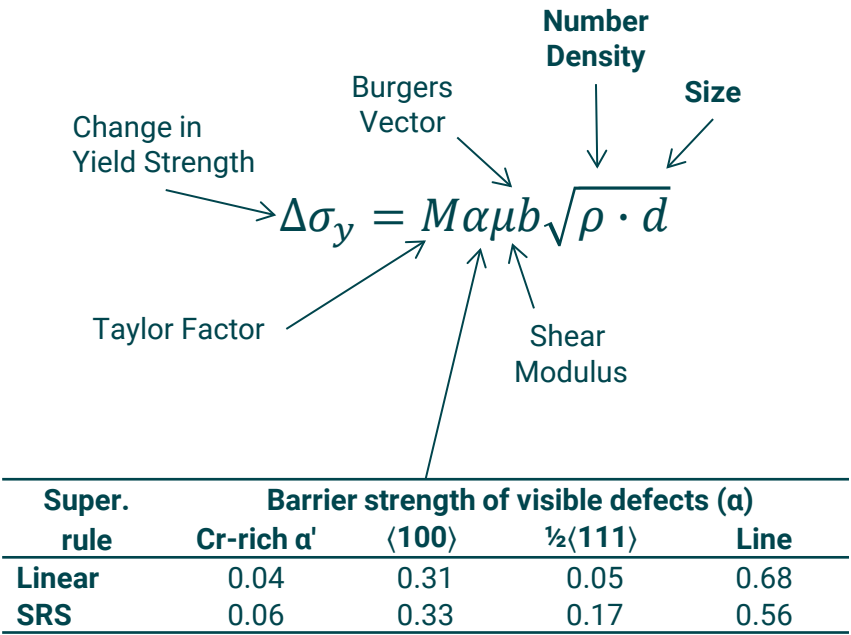
↑

$$[\Delta\rho_{nucl}^2 + \Delta\rho_{mag}^2 \sin^2 \alpha]$$

**Can exploit magnetization
to extract composition**



Radiation tolerance of FeCrAl alloys is analogous to FeCr alloys under similar irradiation conditions



- Dispersed barrier hardening (DBH) has linked radiation-hardening in FeCrAl alloys to formation of Cr-rich α' and dislocation loops after neutron irradiation to 1.8 dpa at 382°C

Need exists to understand the role of composition and temperature on the formation and progression of Cr-rich α' in FeCrAl alloys

A large amount of FeCrAl samples have been irradiated and/or tested over the past 5 years...



...how do we characterize α' from these efficiently?

Magnetic Shielded-SANS measurements:

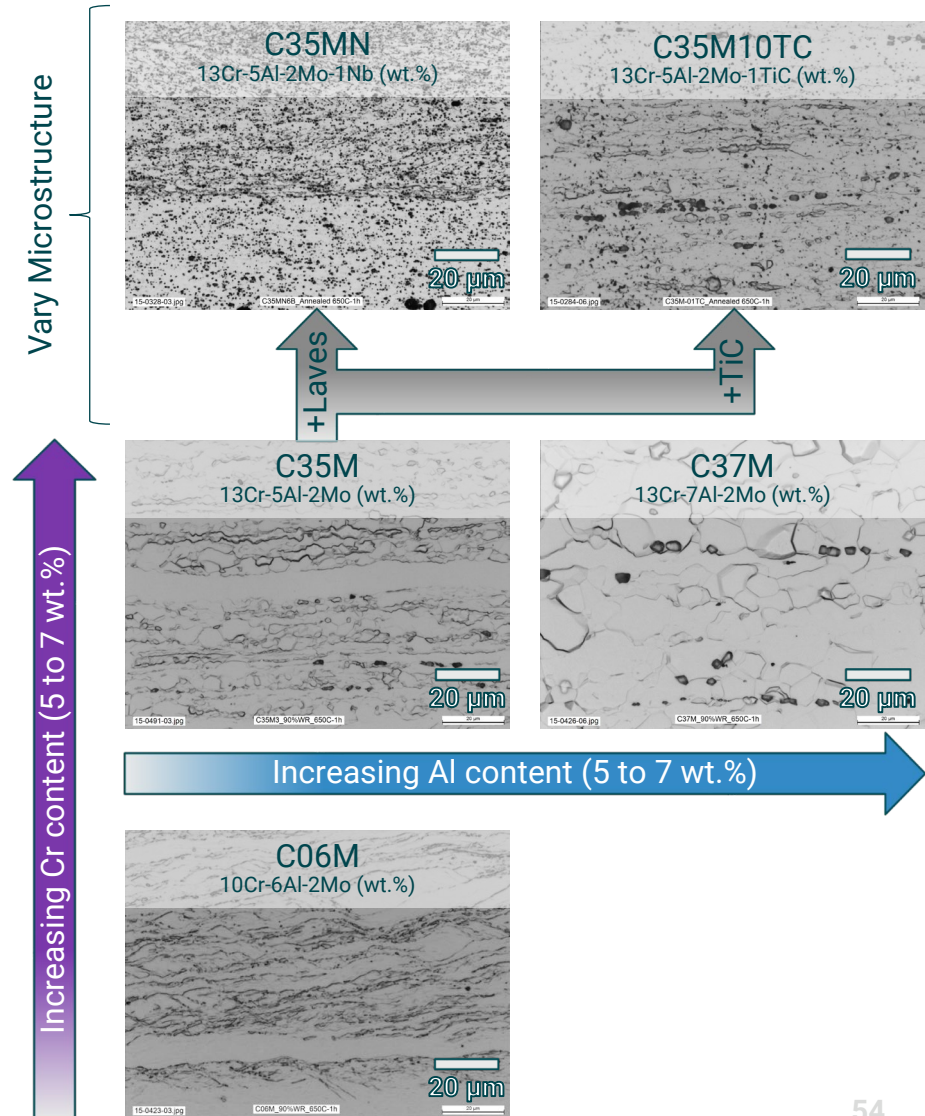
Role of temperature and composition on α'

- Selected FeCrAl alloys irradiated in HFIR to determine temperature, composition, and microstructure trends

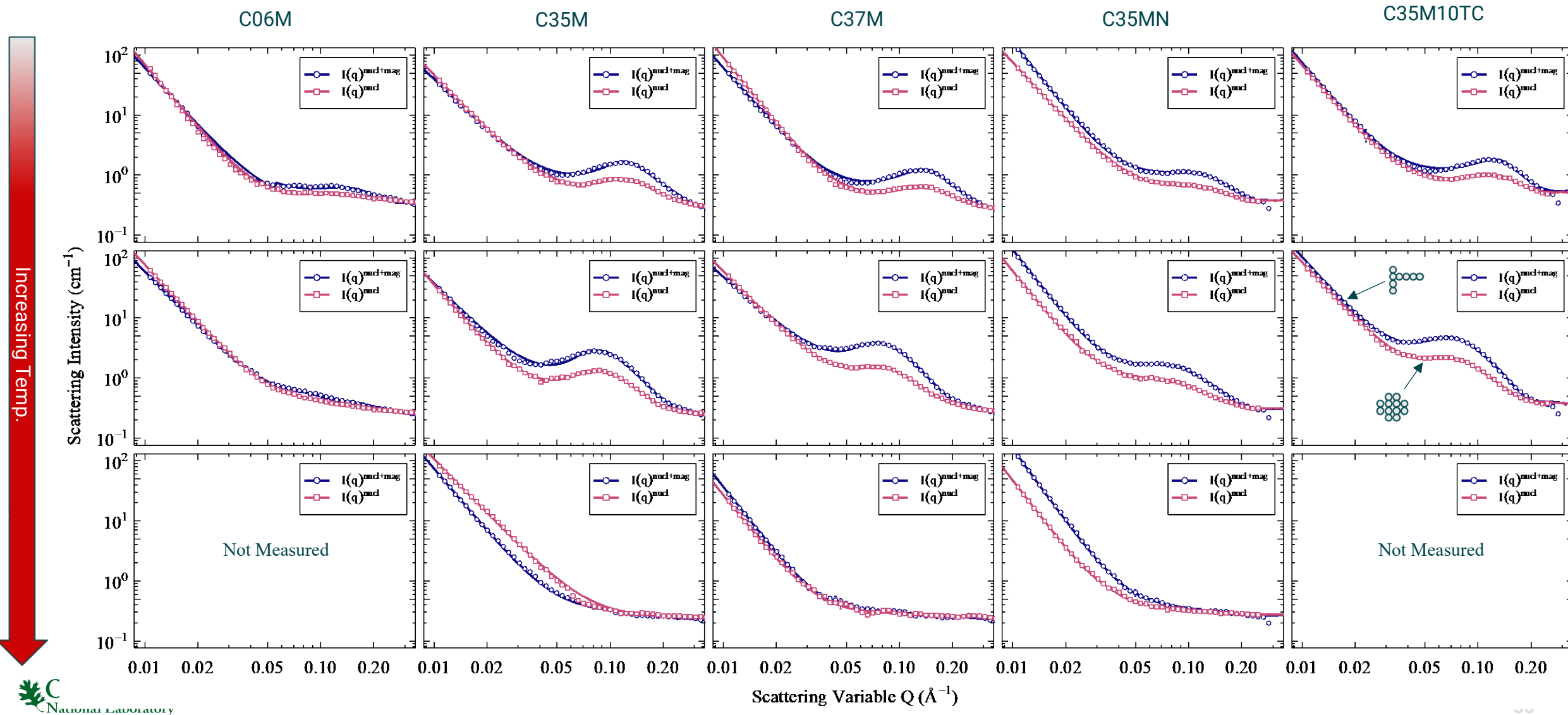
Capsule ID	Number Samples	Neutron Flux (n/cm ² s) E > 0.1 MeV	Neutron Fluence (n/cm ²) E > 0.1 MeV	Dose Rate (dpa/s)	Dose (dpa)	Irradiation Temperature (°C)
FCAT-01	45	1.10×10^{15}	2.17×10^{21}	9.6×10^{-7}	1.9	194.5 ± 37.9
FCAT-02	45	1.04×10^{15}	2.05×10^{21}	9.1×10^{-7}	1.8	362.7 ± 21.2
FCAT-03	45	1.10×10^{15}	2.17×10^{21}	9.6×10^{-7}	1.8	559.4 ± 28.1

- Measurements performed at CG-2 general purpose SANS beamline at HFIR
- Data collected on broken tensile heads in magnetic shielded SANS configuration

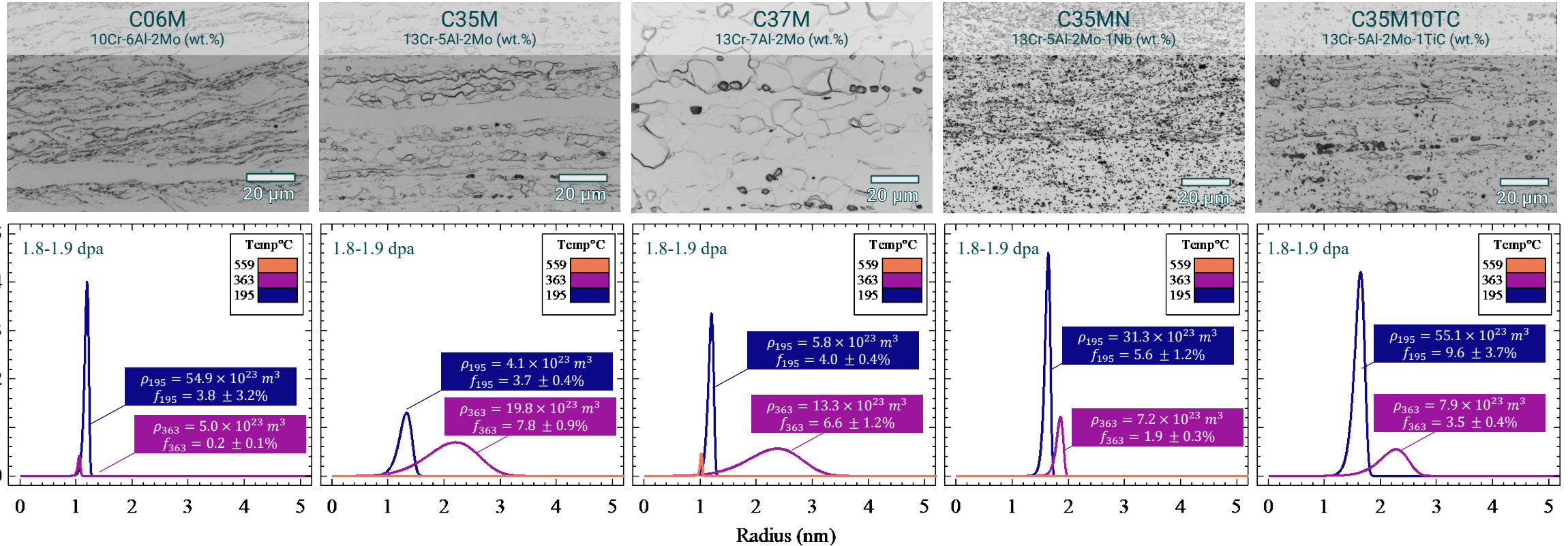
Increasing Temp.



Magnetic Shielded-SANS measurements: Role of temperature and composition on α'



Magnetic Shielded-SANS measurements: Role of temperature and composition on α'



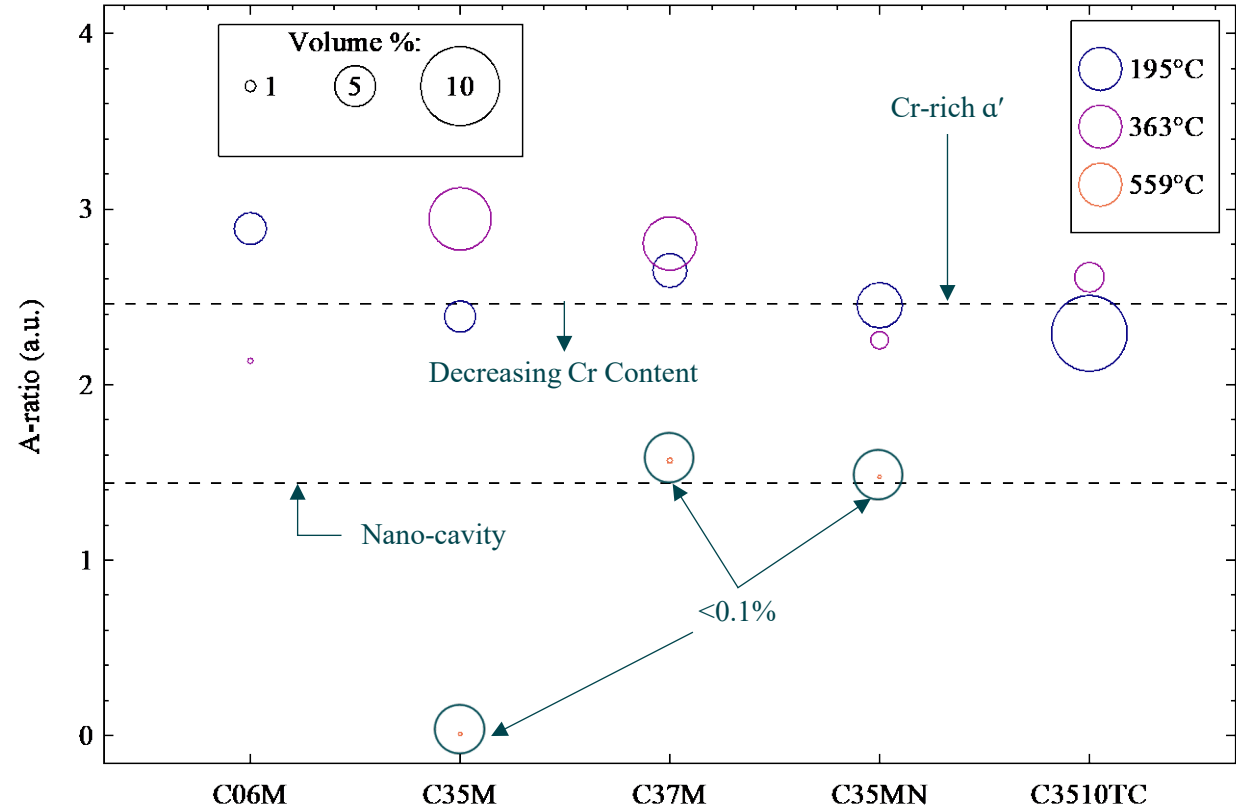
- Increasing irradiation temperature increases mean radius and distribution width
- Cr stronger than Al in control in α' precipitation
- Microstructure (precipitates/grain size) can play a role in α' precipitation

Magnetic Shielded-SANS measurements: Role of temperature and composition on α'

$$\frac{d\sigma}{d\Omega}(q) = \Delta\rho^2 \int_0^\infty \phi(q, R)^2 ...$$

$$[\Delta\rho_{nucl}^2 + \Delta\rho_{mag}^2 \sin^2 \alpha]$$

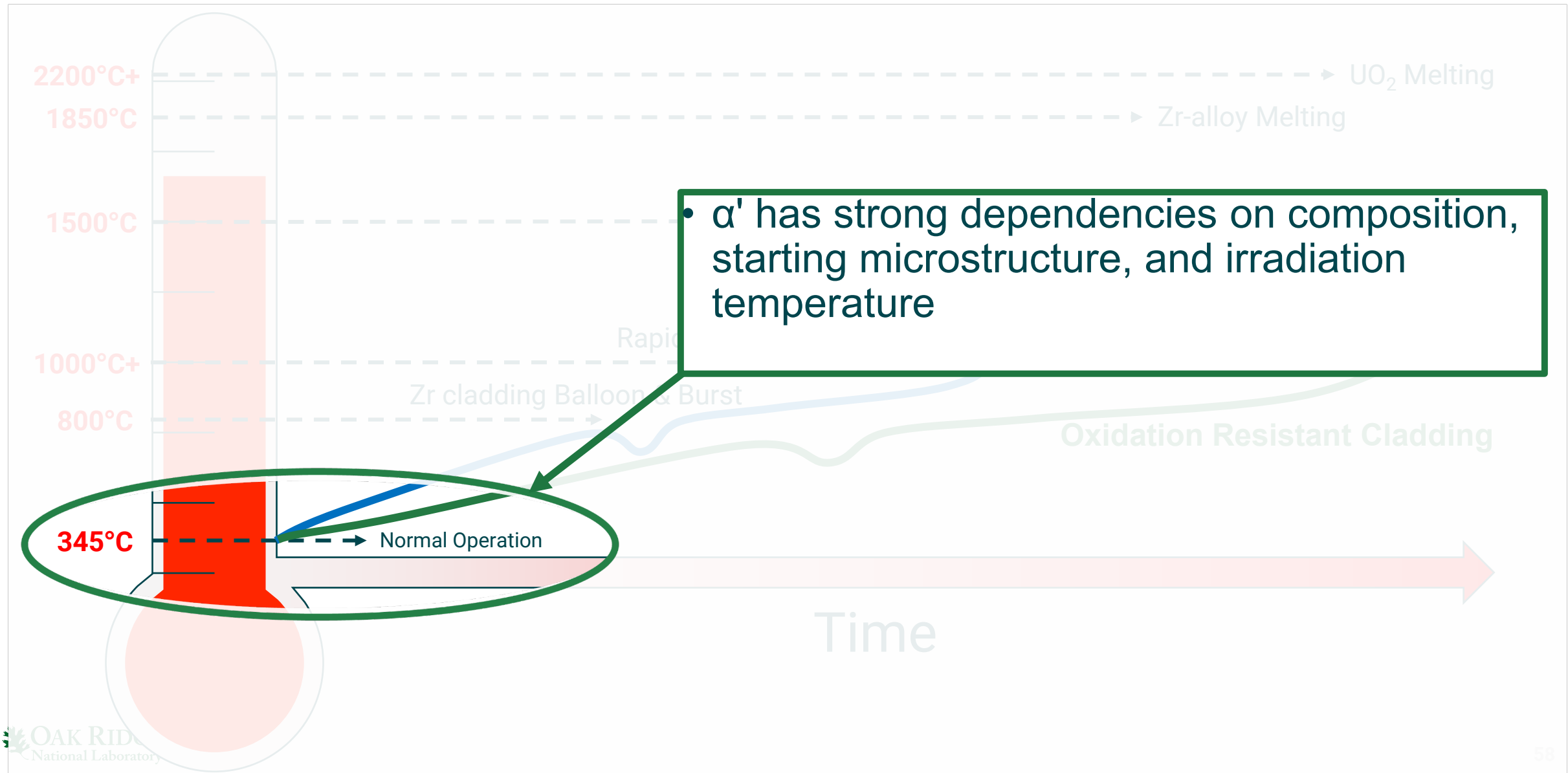
$$A - ratio = \frac{\Delta\rho_{mag+nucl}^2}{\Delta\rho_{nucl}^2}$$



- Magnetic shielded SANS reveals that irradiations below 500°C form primarily α'
 - Typically lower Cr content in 195°C irradiations
- Above 500°C, scattering could be from nano-cavity formation

Practical Application of Ex-situ SANS results:

α' concern for normal operation, but can be tuned

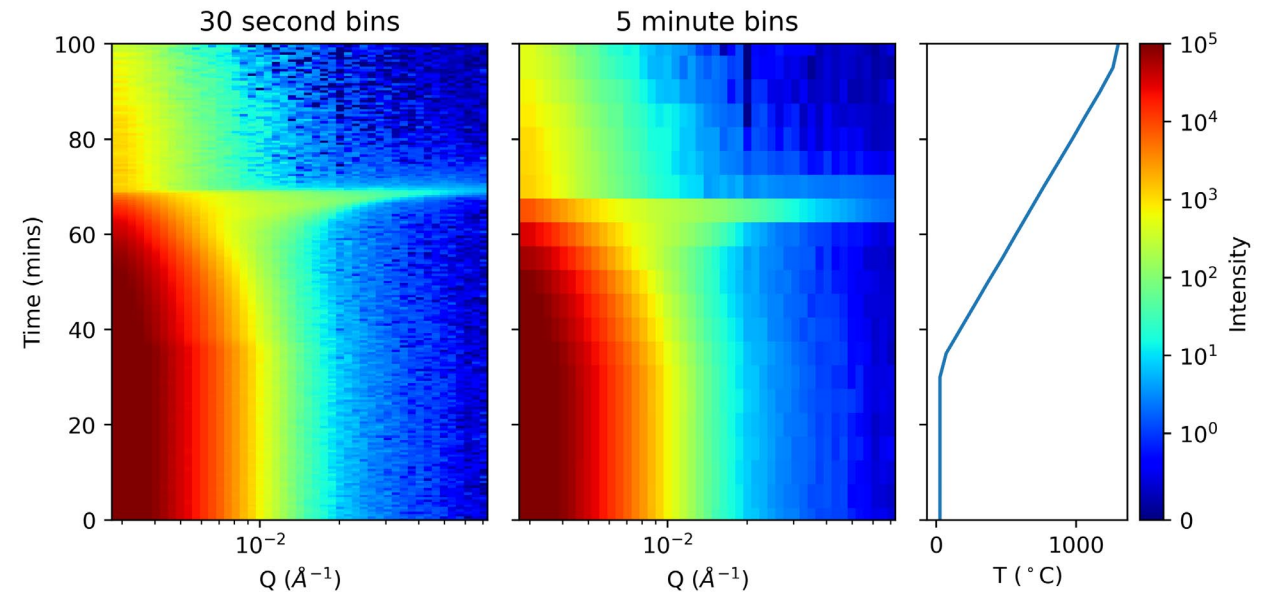
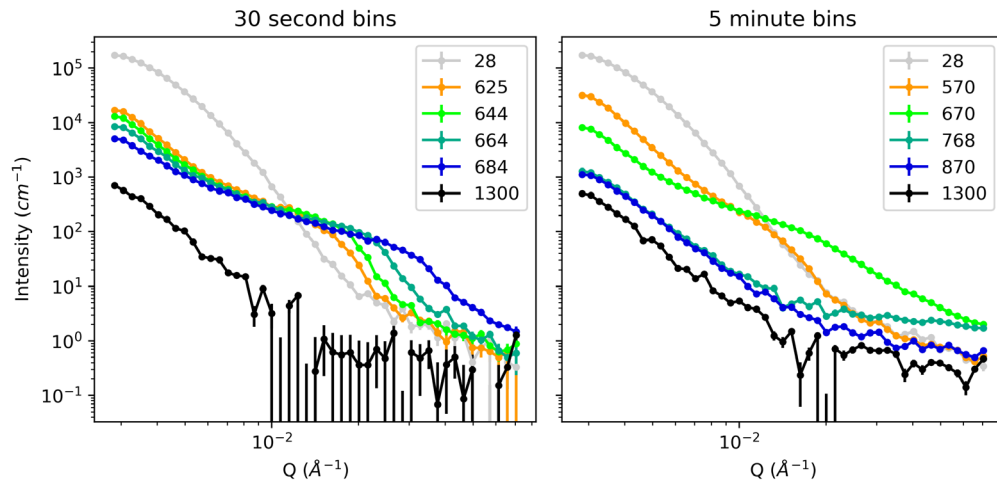


New Directions with SANS

Time-resolved SANS

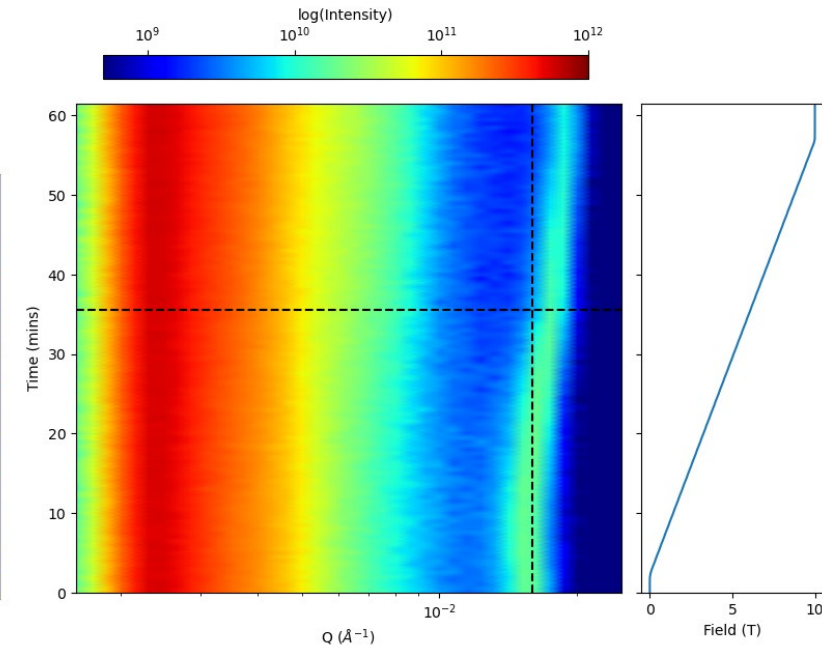
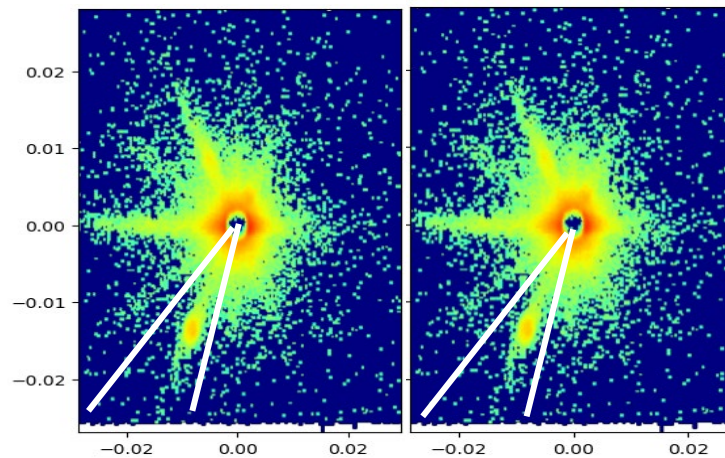
GI-SANS

Time-resolved SANS (TR-SANS)



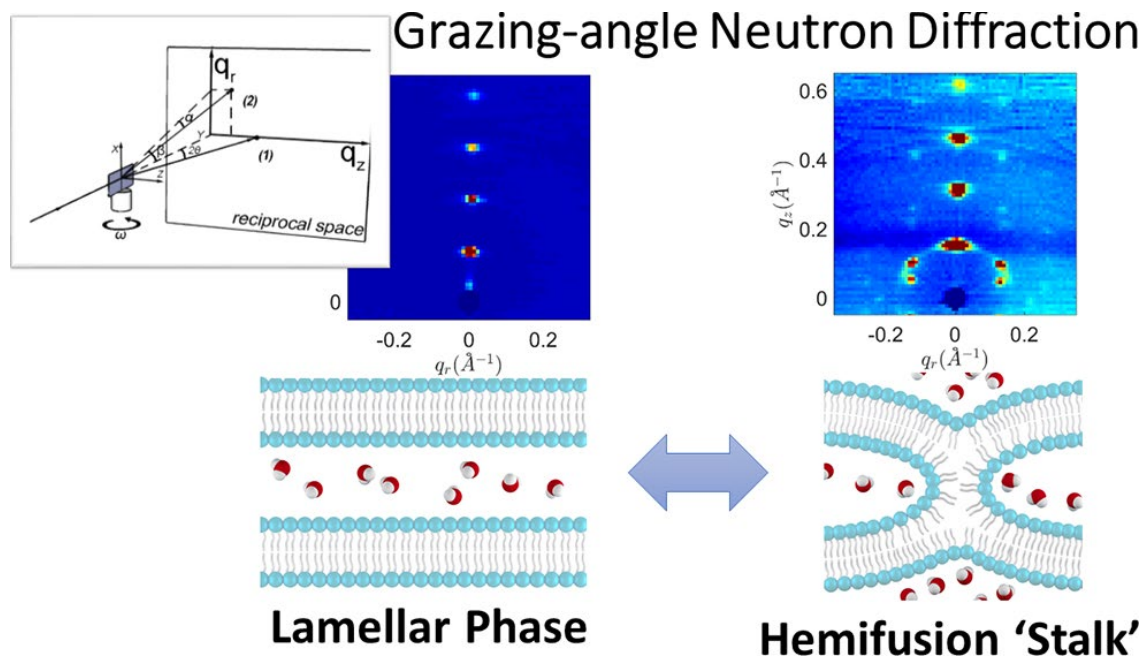
- Additively manufactured reactor steels
- Data(above) collected during ramping temperature in a vacuum furnace.
- Post-processing software can re-bin the data either by time or log value(magnetic field)

- Data collected during magnetic field ramp from 0 to 10 T
- Data sliced by 30 second slices as the field is ramped.



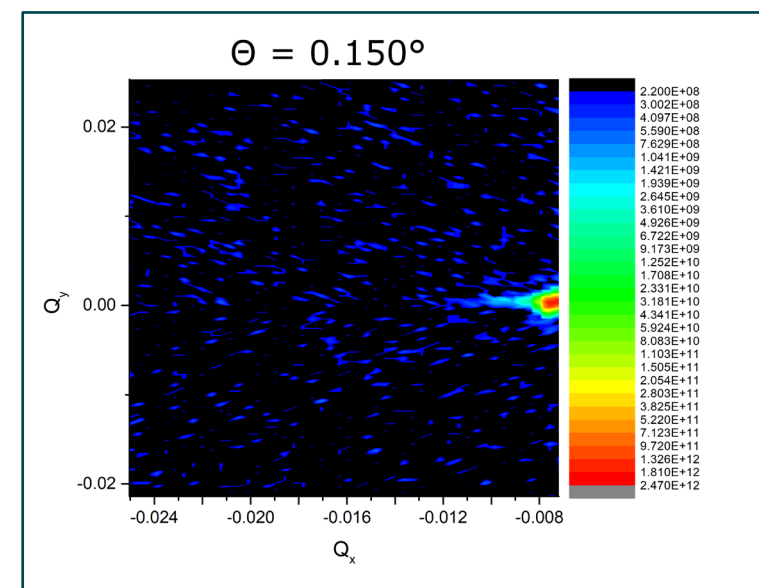
GI-SANS

- Being developed at ORNL at EQ-SANS but GP-SANS, BIO-SANS can do this technique.
- Measuring the specular and off-specular versus sample angle encodes both in-plane and out-of-plane structure of the systems.



S. Qian and D. K. Rai J. Phys. Chem. Lett. 2018, 9, 5778-5784

Fe/Gd multilayers measured with a large (195 mT) out-of-plane field in the skyrmion phase



WLNC Liyanage *et. al* Phy. Rev. B **107**, 184412 (2023)

Experts at ORNL on GI-SANS:

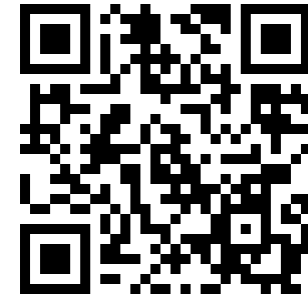
- William Heller
- Changwoo Do
- Valeria Lauter



Neutron Scattering
Society of America

Thank you for your attention.

- Probes bulk structures from 0.1 nm – 1 μ m
- Contrast tuning with isotopes & magnetism
- Works with diverse sample environments
- Applies across soft matter, magnetism, and alloys



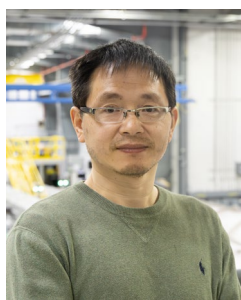
ORNL neutron website

Instrument Teams:

GP-SANS



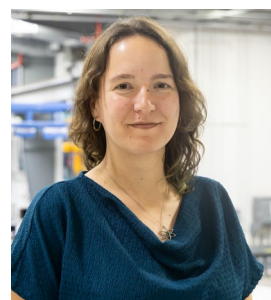
Lisa
DeBeer-Schmitt



Lilin
He



Karin
Bichler



Arina
Rostopchina

Bio-SANS



Venky
Pingali



Wellington
Leite



Volker
Urban



Felicia
Gilliland

USANS



Wei-ren Chen



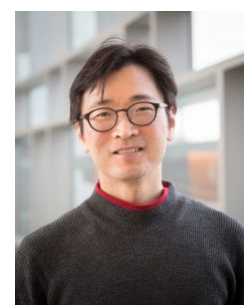
Gernot Rother



Yingrui Shang



Carrie Gao



Changwoo Do



William Heller



Gergely Nagy

EQ-SANS

NXS Lecture - Lisa DeBeer-Schmitt

"Small Angle Neutron Scattering"



Extra slides



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