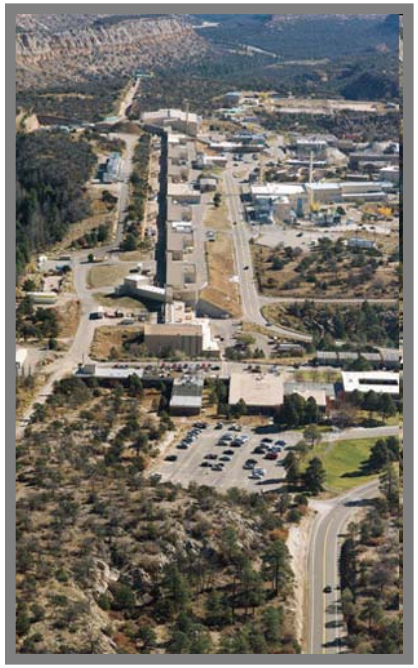
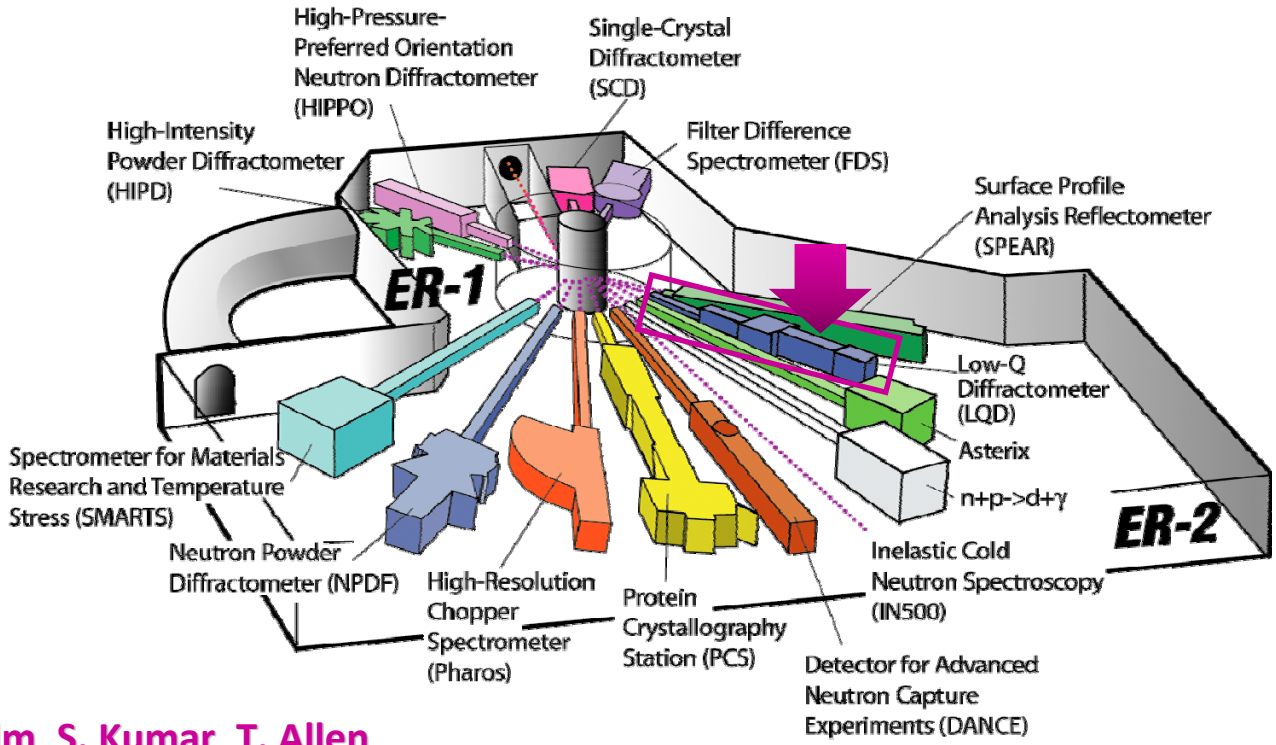
LQD measurements on ATR's irradiated ferritic-martensitic steels

Los Alamos NATIONAL LABORATORY EST. 1943



Lujan Center at LANSCE



M. H. Braga, M. Hartl, R. Hjelm, S. Kumar, T. Allen

Neutron Scattering Advantages:

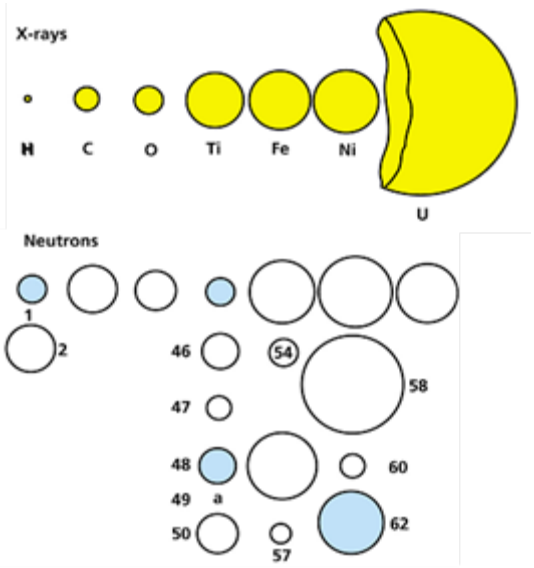
- Neutrons are highly penetrating; bulk materials can be studied.

- Neutrons exhibit different interactions with varying isotopes of the same element.

- The difference in scattering offers the unique capability of labeling
e.g. the exchanging of hydrogen against deuterium.

- There is a scattering length associated with the interaction of neutrons with a magnetic moment; magnetic structure can be studied.

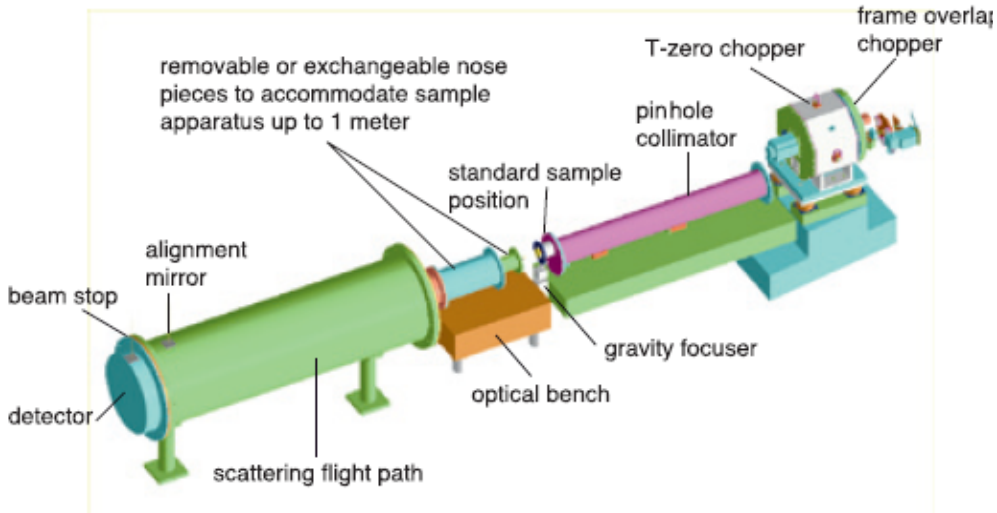
X-ray and neutron scattering lengths for selected elements



LANSCE can receive radioactive samples

Small Angle Neutron Scattering with LQD

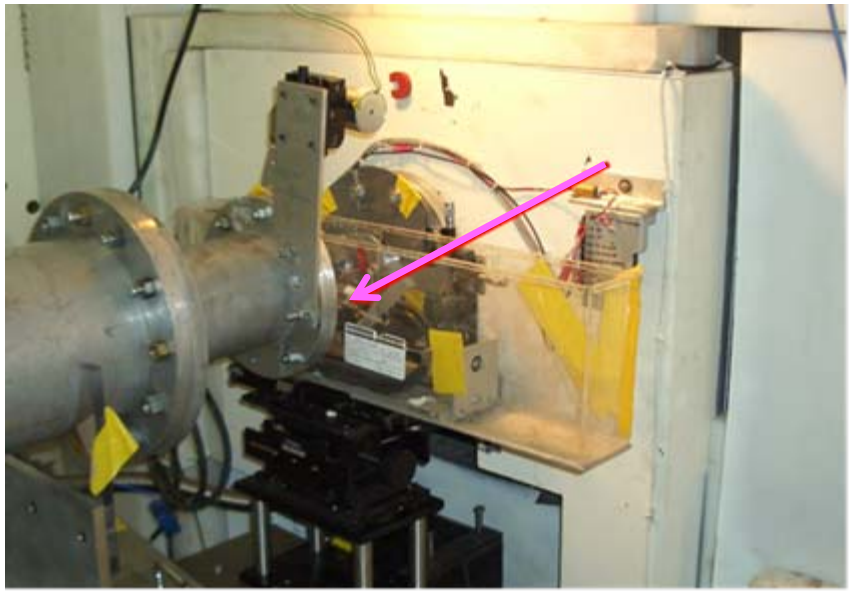
Instrument Scientists: Rex Hjelm (hjelm@lanl.gov) and Monika Hartl (hartl@lanl.gov)

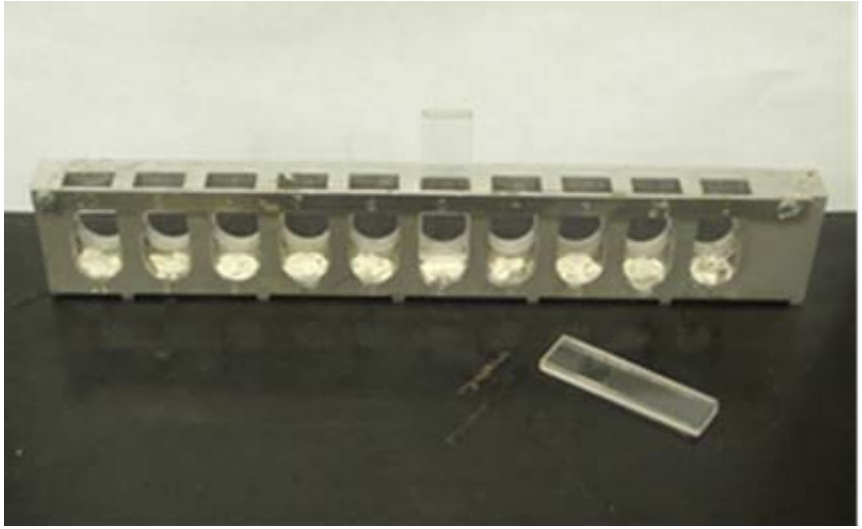


Q range: **0.003** to **0.5** $Å^{-1}$
Source to sample: 8.40 m
Sample to detector: 4.32 m
Intense source of long wavelength ("cold") neutrons; λ=1-16 Å
Energy and wavelength vary as TOF
~1 nm – 100 nm features

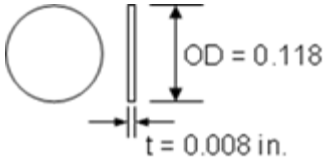
The low-Q diffractometer, LQD, is designed to study large-scale structures with dimensions from 10 Å to 1000 Å (1 nm – 100 nm). Examples of problems that LQD can help solve include phase separation, phase morphology, and critical phenomena in hard and soft matter and in magnetic structures, colloid and polymer structure, biomolecular organization, and bubble formation in metals.

LQD's sample holders (cuvettes)





Sample area of LQD with the sample changer in place. The sled (pink arrow) holds between 5 and 10 samples.

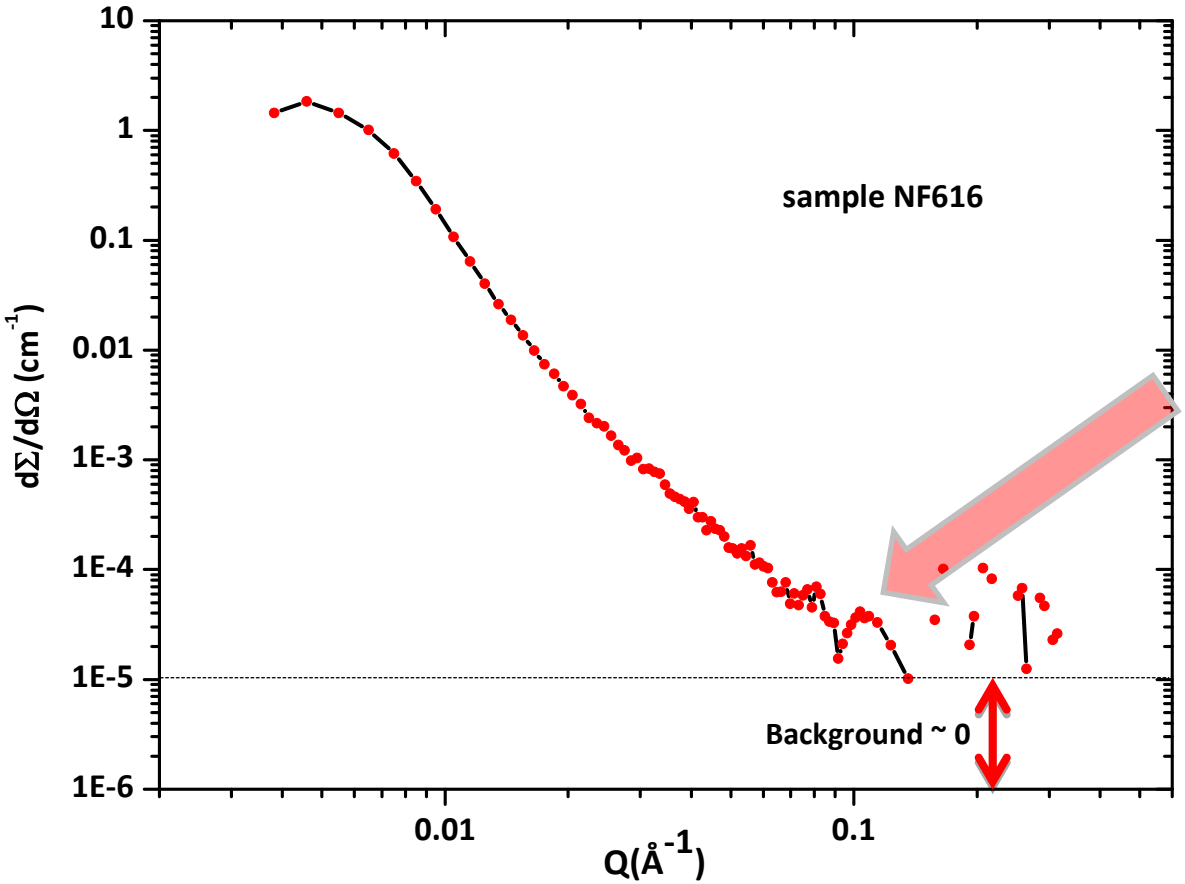


Shape and dimensions of our samples

Cuvettes are easy to fill with solids as well as liquids. The thickness of the cell is 1 or 2 mm; Special cells with 3mm and 5mm thickness are also available.

NF616

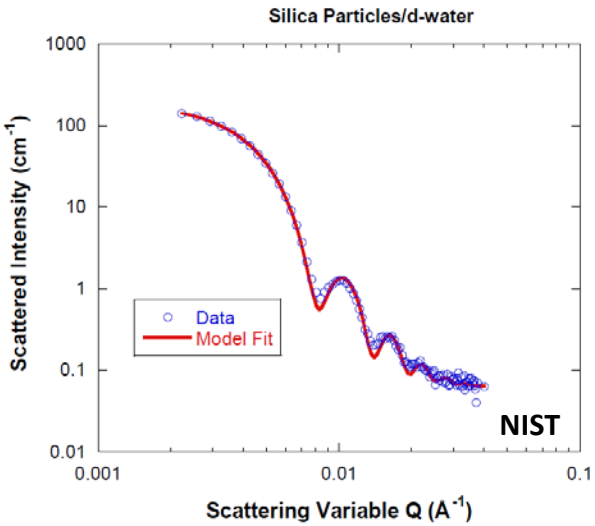
Material ID	Density (g/cm^3)	B	C	N	O	Al	Si	P	S	Ti	V	Cr	Mn	Fe	Ni	Cu	Nb	Mo	W	Other
HCM12A	7.7	-	0.09	0.0424	-	-	0.30	0.005	-	-		10.50	0.52	bal	1.04	1.5	-	0.26	1.43	Nb+Ta: 0.05
NF616	7.7	0.0017	0.109	0.0474	0.0042	0.005	0.102	0.012	0.0032	-	0.194	8.82	0.45	bal	0.174	-	0.064	0.468	1.87	



Irradiation to 3 dpa

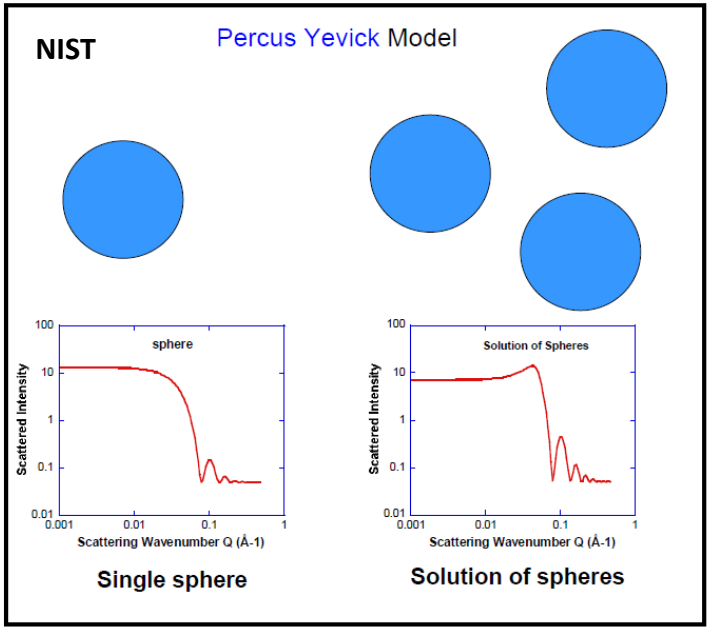
The effect of polydispersity is seen to broaden the peaks

Black line is just a guide for the eyes

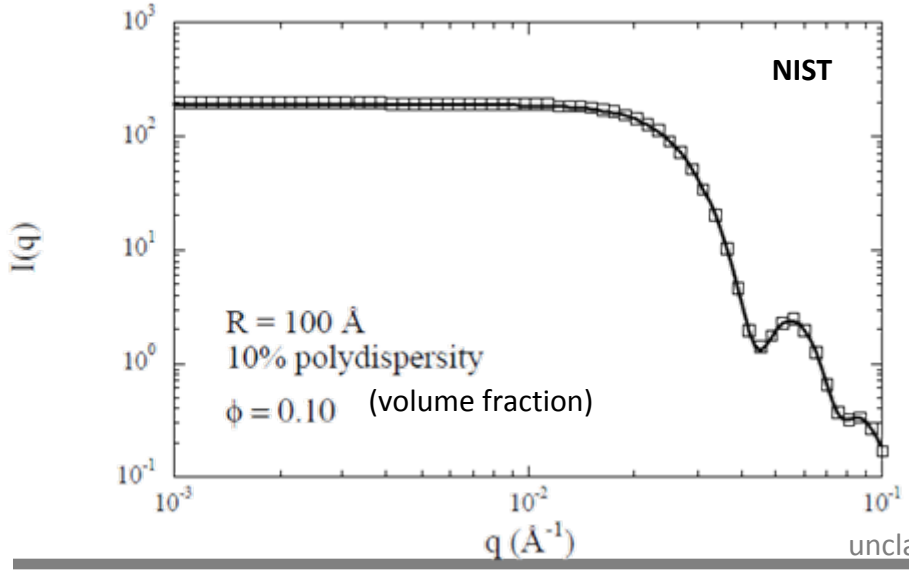


The case of the dilute solutions is characterized by scattering from "isolated" particles. A 0.1% solution of silica particles in d-water is shown here. The higher order peaks are a signature of monodispersity and are limited by instrumental resolution. The fitted radius is R = 563 Å.

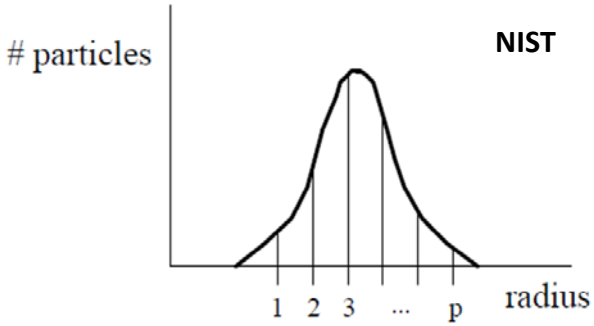
Solution of Spheres



Model calculations for polydisperse spheres



polydisperse spheres



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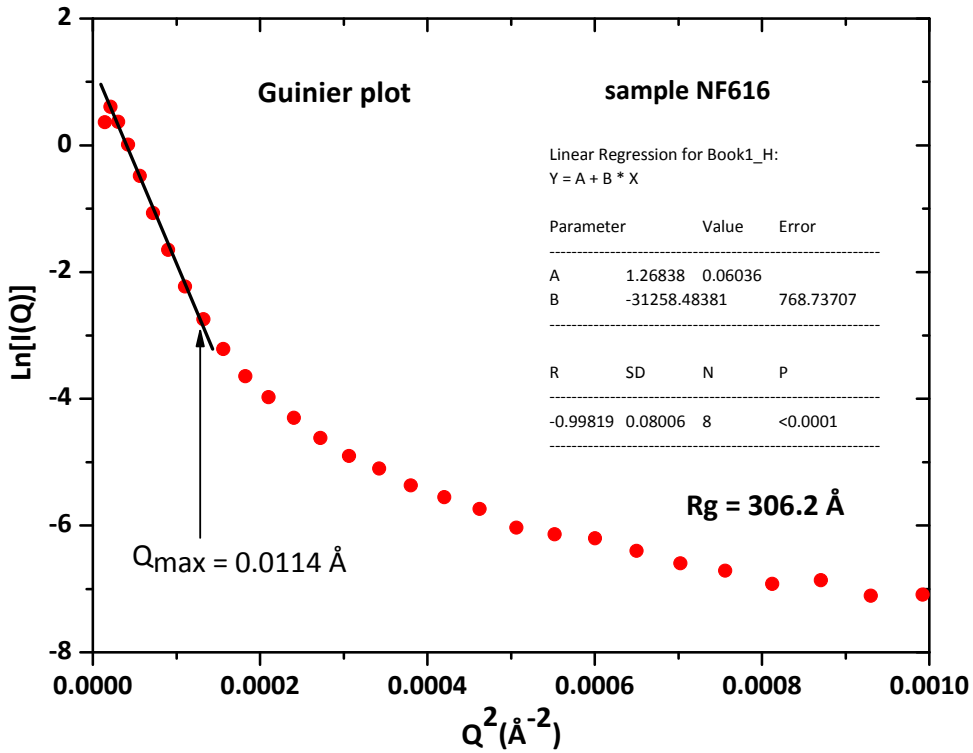
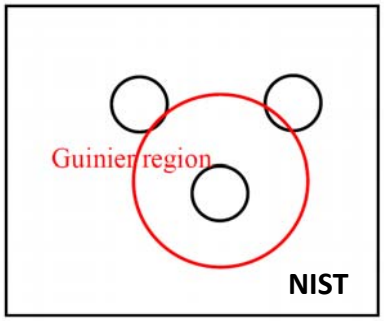
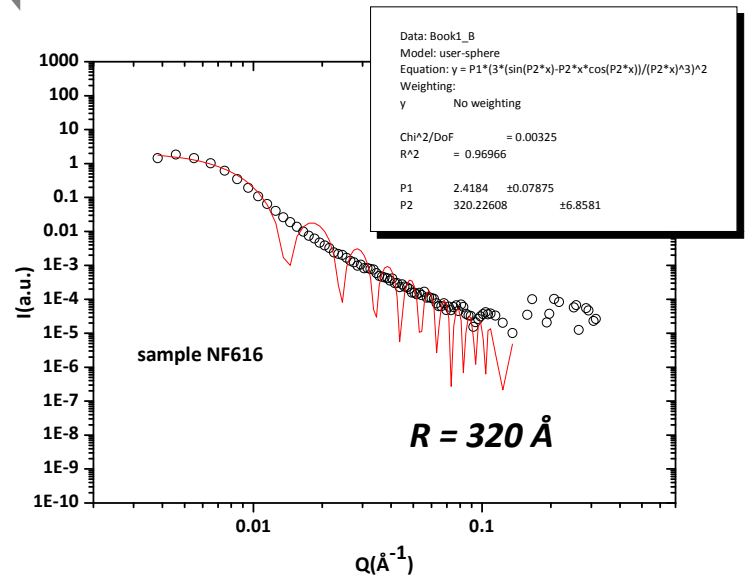




$$R^2_g = \frac{3}{5} R^2$$

R = 395.3 Å ~ 40 nm



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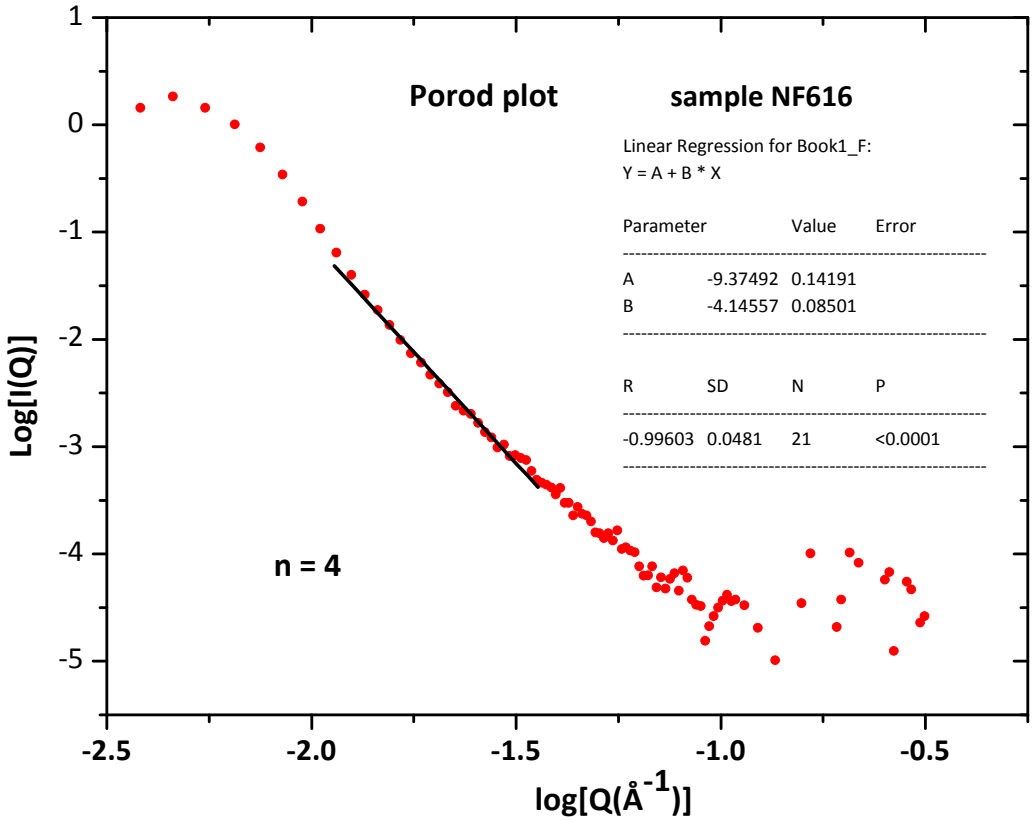

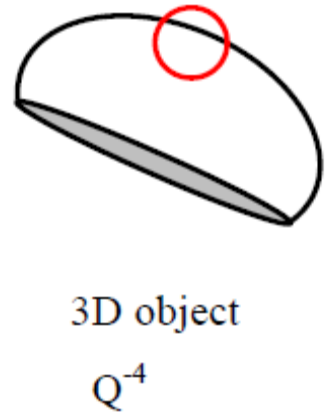

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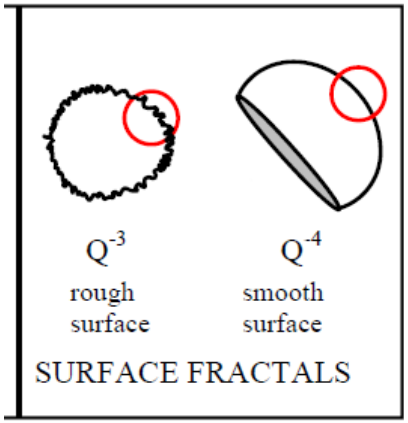

$$I = \frac{A}{Q^n} + B \text{ or } Log[I(Q)-B]=Log(A)-nLog(Q)$$

HCM12A

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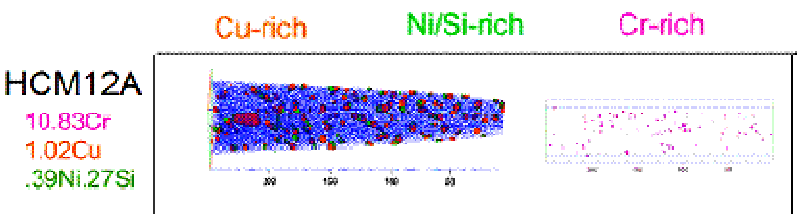
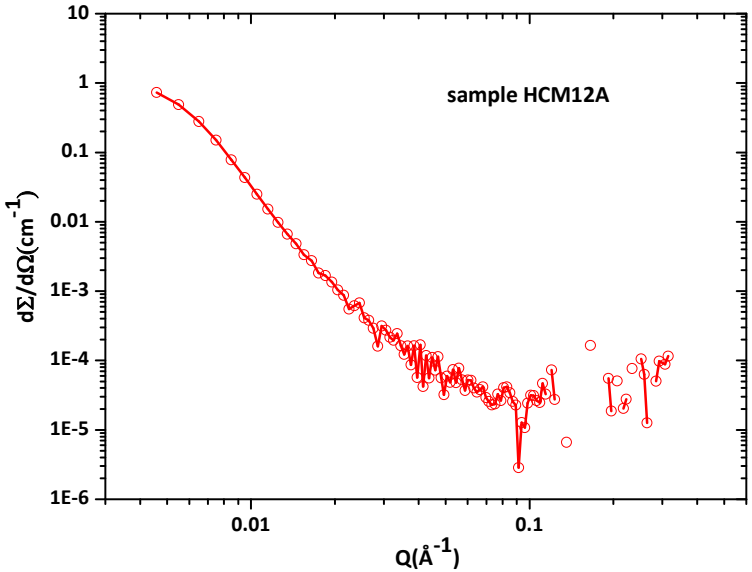
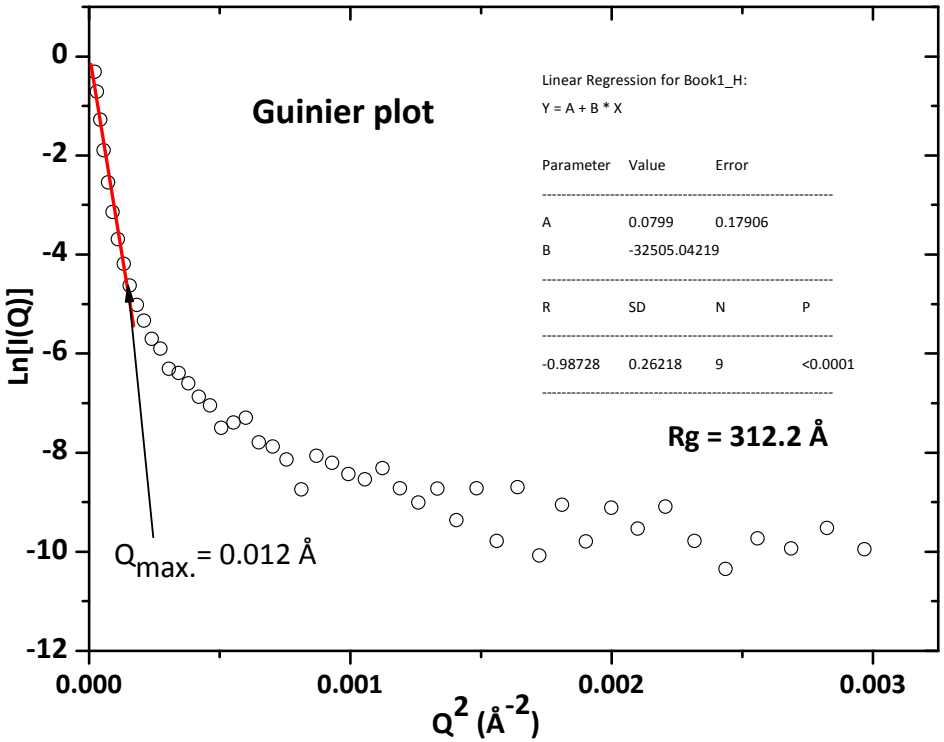


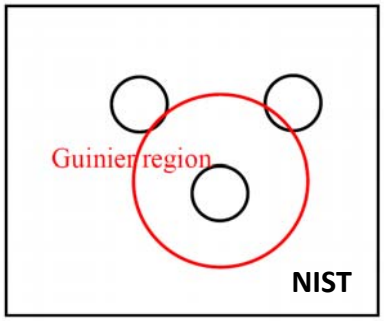
Irradiation to 3 dpa

*Precipitates formed in HCM12A F-M alloys following irradiation to **7 dpa** at 400°C.*



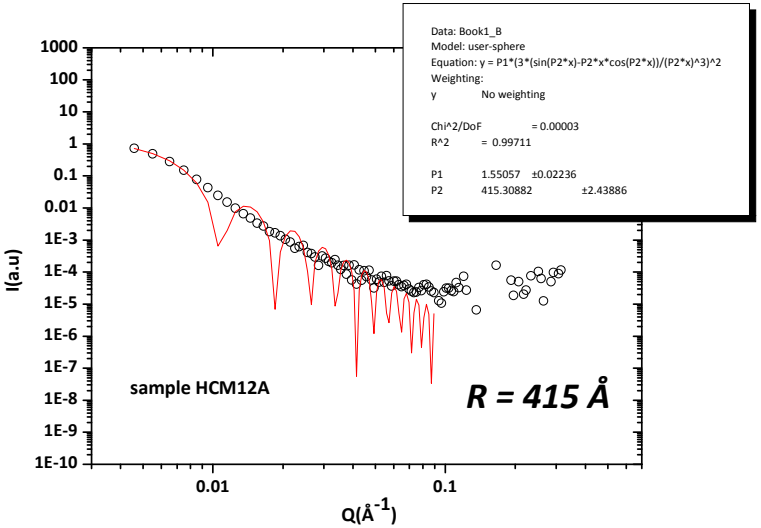
HCM12A





$$R^2{}_g = \frac{3}{5} R^2$$

R = 403.0 Å ~ 40 nm



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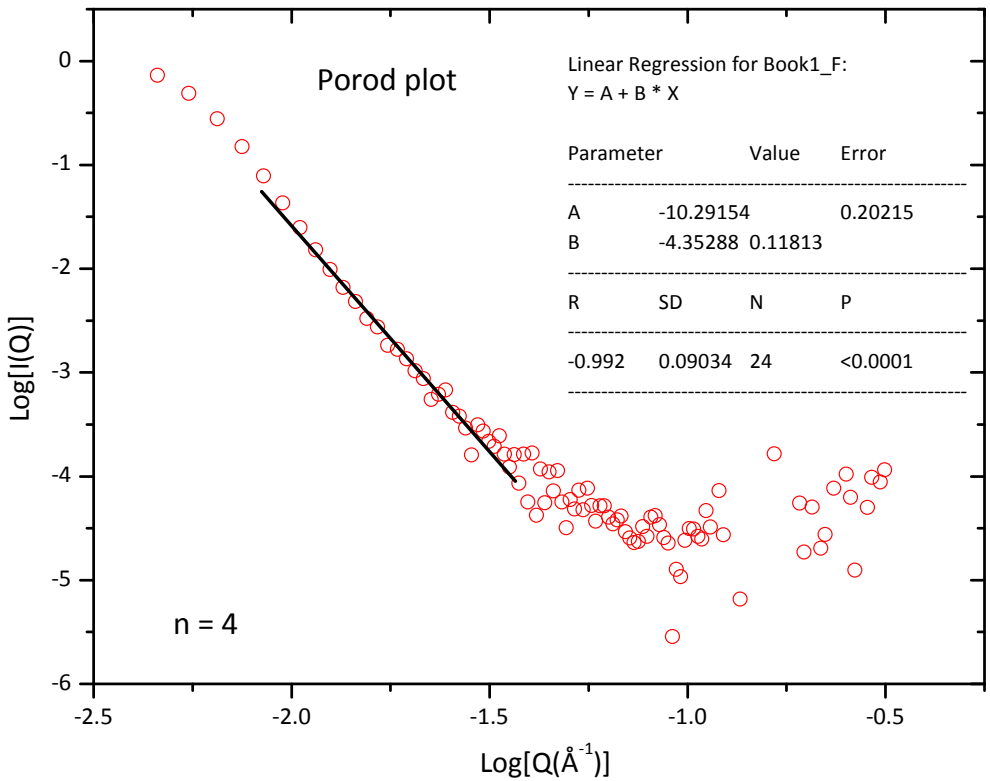

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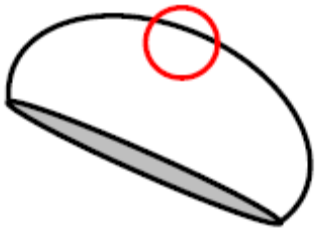
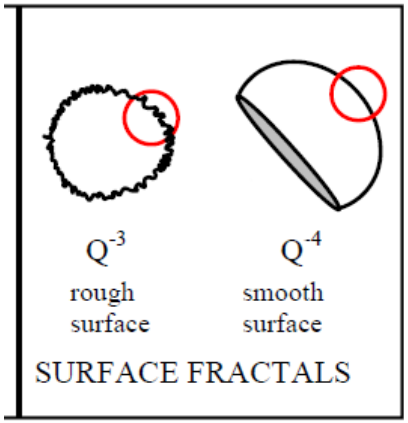


3D object

Q^{-4}

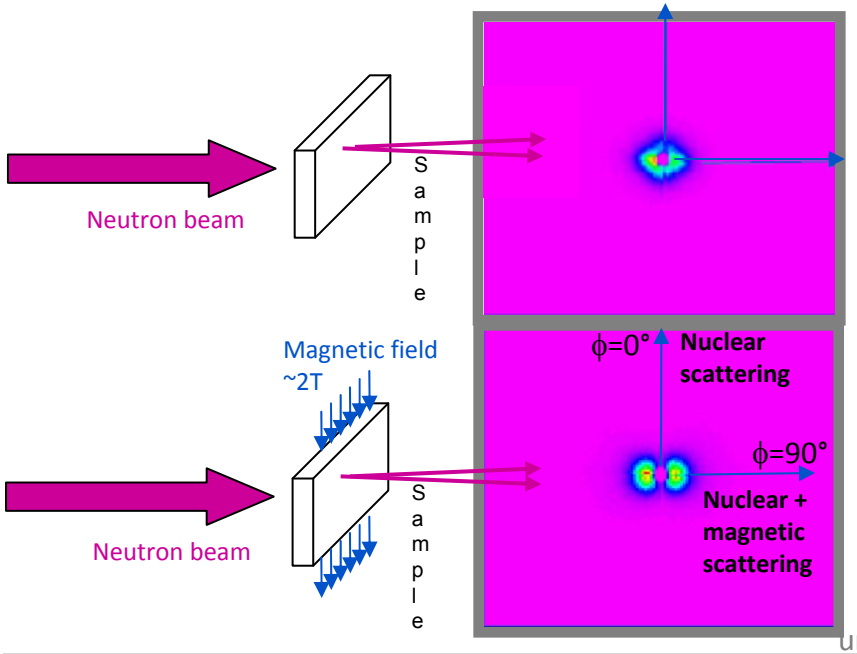
NIST



Magnet

A-ratio scattered intensities perpendicular and parallel to the sample magnetization.
For chemically and magnetically homogeneous particle, it is given by (assuming that the form factor P(Q) is the same for nuclear and magnetic contributions):

$$A = \left(\frac{d\Sigma}{d\Omega}\right)_{\perp H} \Big/ \left(\frac{d\Sigma}{d\Omega}\right)_{// H} = 1 + \left(\frac{\Delta\rho_{mag}}{\Delta\rho_{nucl}}\right)^2$$



Little-no contrast between matrix and particle.

Particles and matrix have different magnetic properties.

SANS experimental setup for measuring the contrast between matrix an particles in a magnetic field. In the case of LQD (measuring in transmission geometry), only the component normal to the sample plane contributes to b_{magn}.

SANS with Magnetic Contrast (unpolarized neutrons)

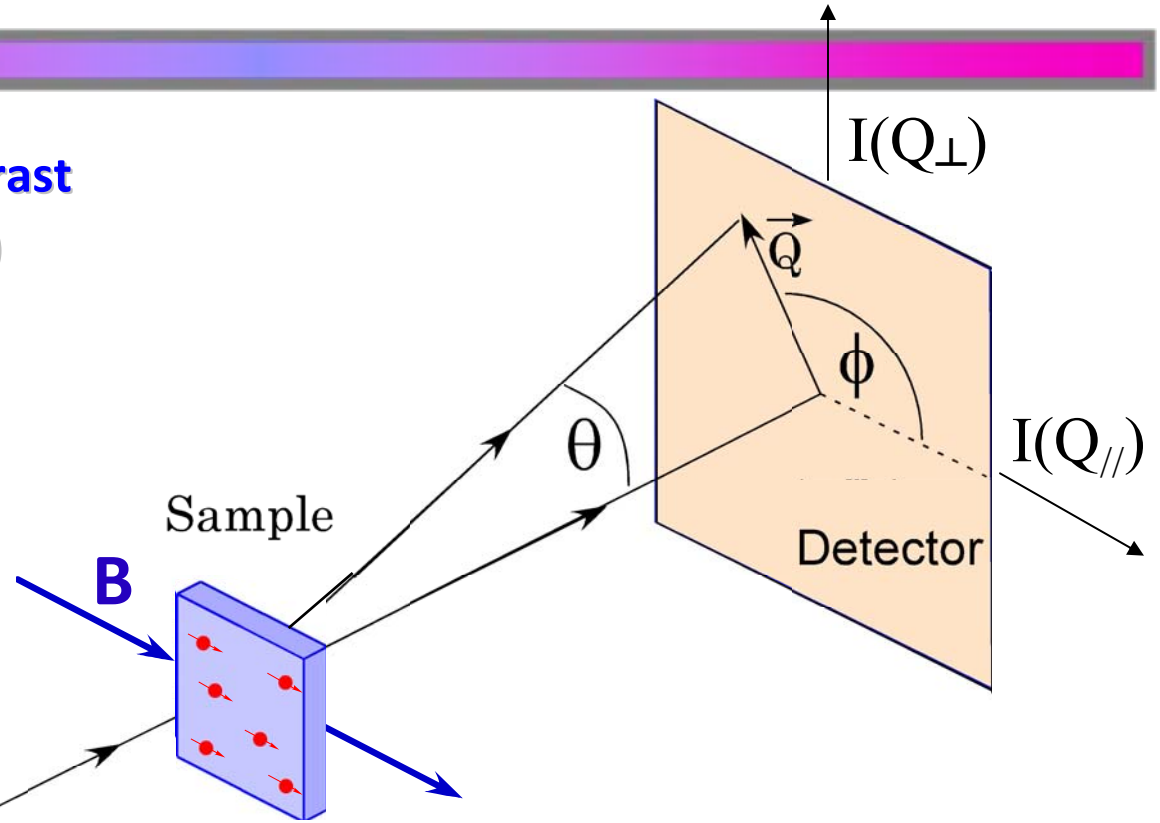
Magnetic contrast

$$(\Delta M)^2 = (M_p - M_o)^2$$

Nuclear contrast

$$(\Delta\rho)^2 = (\rho_p - \rho_o)^2$$

"particle" matrix



$$I(Q) \propto (\Delta\rho^2 + \Delta M^2 \sin^2\varphi)\, F^2(Q)\, S(Q) +$$ similar terms for each species

$$\frac{I(Q_\perp)}{I(Q_{||})} = \frac{\Delta\rho^2 + \Delta M^2}{\Delta\rho^2}$$

Magnetic scattering comes From $\Delta M \perp Q$ *only*!

$$A = \left(\frac{d\Sigma}{d\Omega}\right)_{\perp H} \Big/ \left(\frac{d\Sigma}{d\Omega}\right)_{//H} = 1 + \left(\frac{\Delta\rho_{mag}}{\Delta\rho_{nucl}}\right)^2$$

$$\frac{d\Sigma}{d\Omega}(q,\phi) = NV^2(\Delta\rho)^2|S(qr)|^2$$
$$= NV^2(\Delta\rho)^2\{3[\sin(qr) - qr\cos(qr)]/(qr)^3\}^2$$
$$\rho^{nuc} = \sum_i \frac{b_i X_i}{V}$$

A-ratio

magnetic scattering length: $b_{magn}(Cr) = 1.08 \cdot 10^{-12}$ cm
$b_{magn}(Fe) = 0.6 \cdot 10^{-12}$ cm

$\Delta\rho$ - contrast factor: difference in scattering length density

Examples (from J. Van den Bosch)

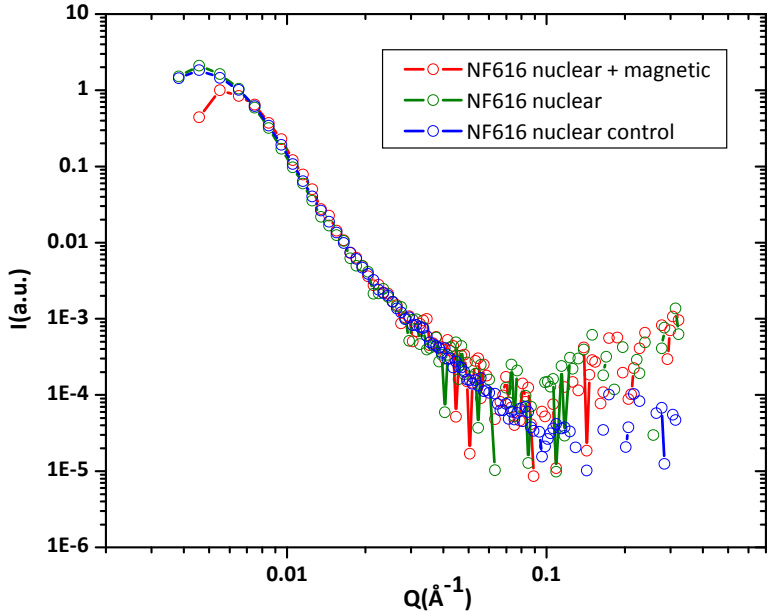
- M23C6 ($Cr_{0.65}Fe_{0.35}$): 3.5

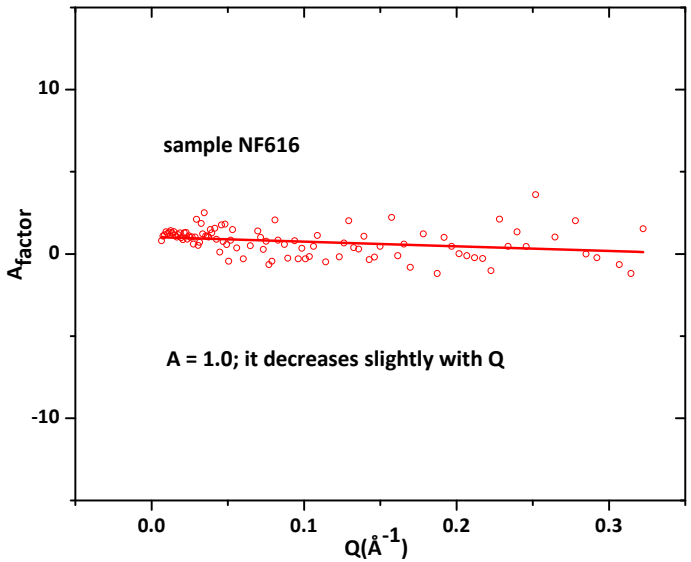
- **Vacancy clusters or voids: 1.0-1.4**

- α' Cr precipitates: 2.03-2.13
(taking into account the average magnetic moment: μ=2.20-2.39 C_{Cr})

- Cr-13Mo-8Fe-3Si: 2.35
(measured by Gelles and Thomas in HT9 following neutron irradiation at 425°C to high dose)

NF616





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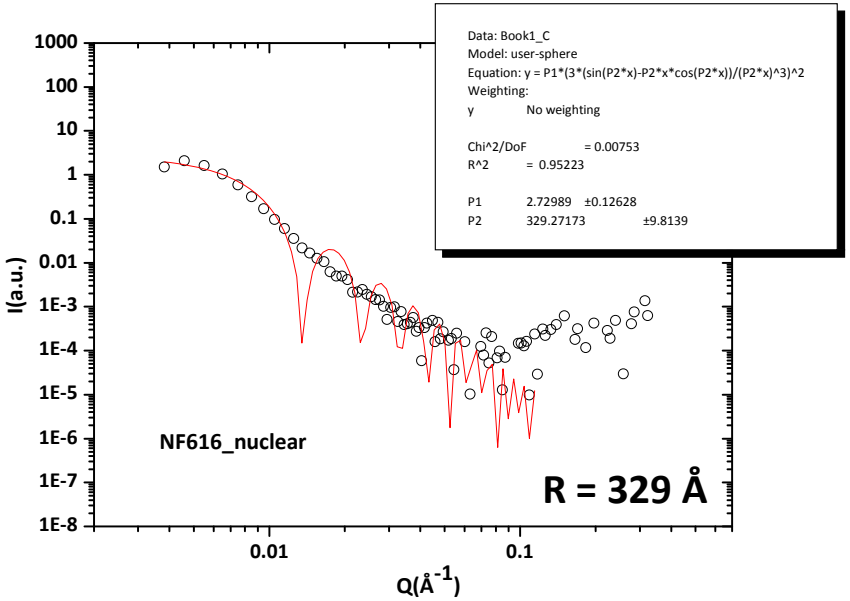

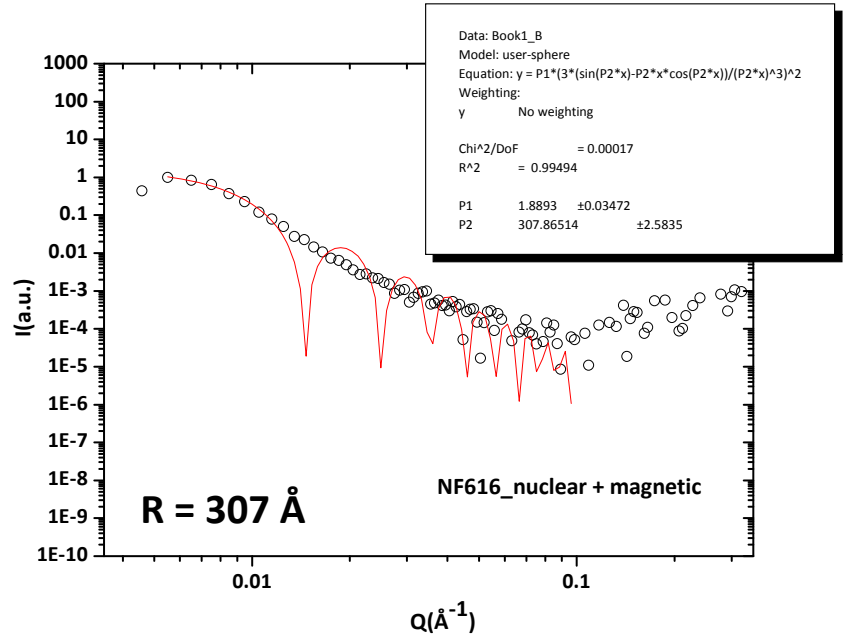

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$$F_{sphere}(Q,R) = \left[\frac{3[\sin(QR) - QR\cos(QR)]}{(QR)^3}\right]^2$$

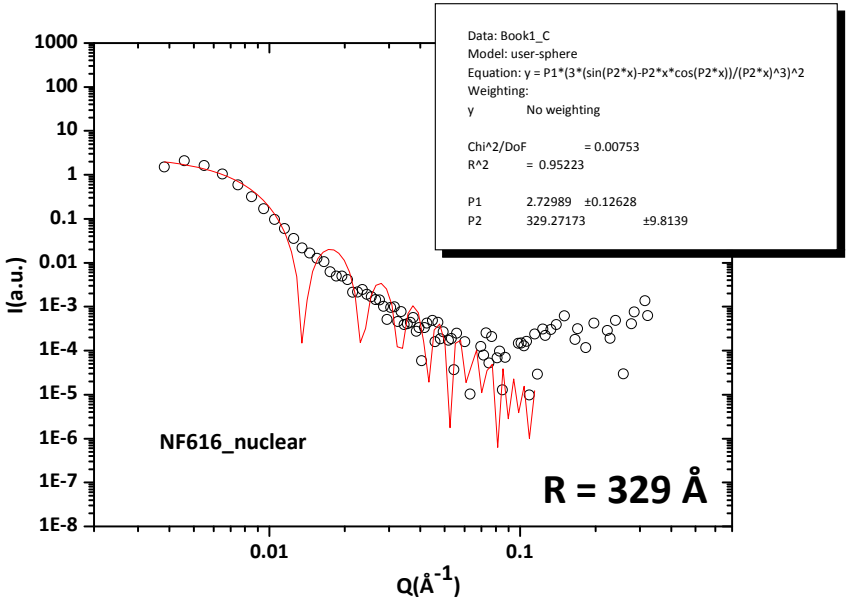
$$A = 1 + \left(\frac{\Delta\rho_{mag}}{\Delta\rho_{nucl}}\right)^2 \cong 1 \Rightarrow (\Delta\rho_{mag} << \Delta\rho_{nucl}) \Rightarrow NV^2(\Delta\rho_{nucl})^2 \cong NV^2(\Delta\rho_{nucl+mag})^2 = P1$$

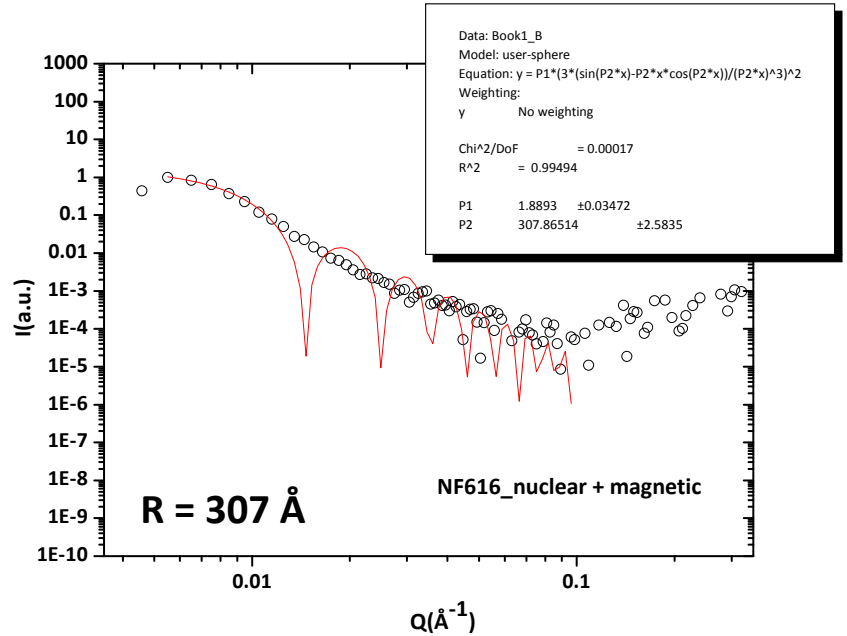
R = 395 Å ~ 40 nm (Guinier) no magnet

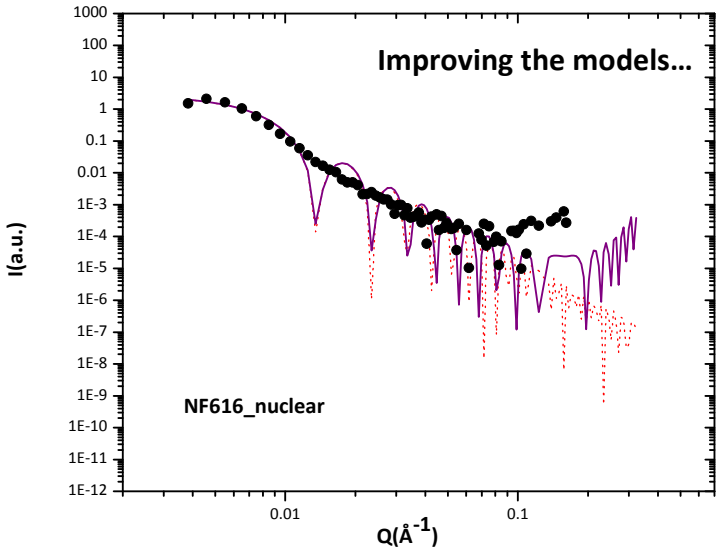
R = 320 Å sphere model no magnet

P1 nuclear = 2.7 cm^{-1}
P1 nuclear + mag = 1.9 cm^{-1}
P1 control = 2.4 cm^{-1}

NF616







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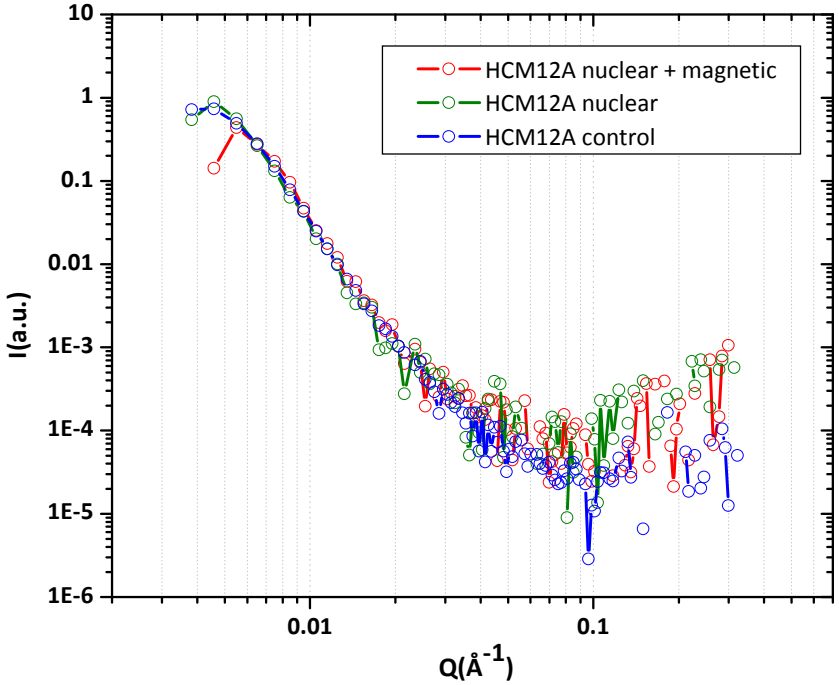
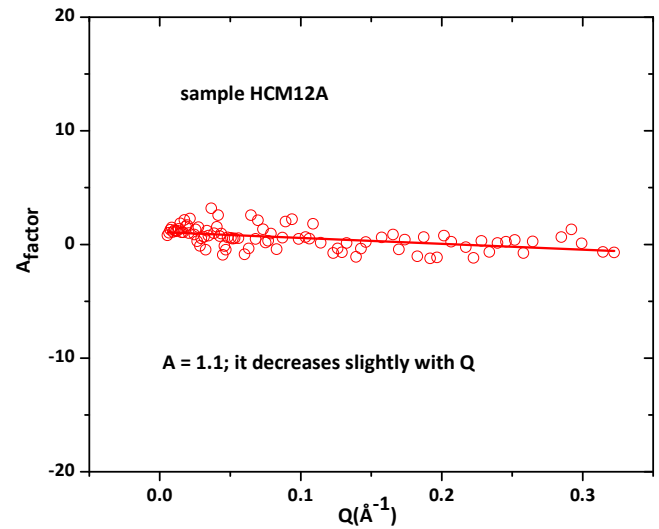
HCM12A





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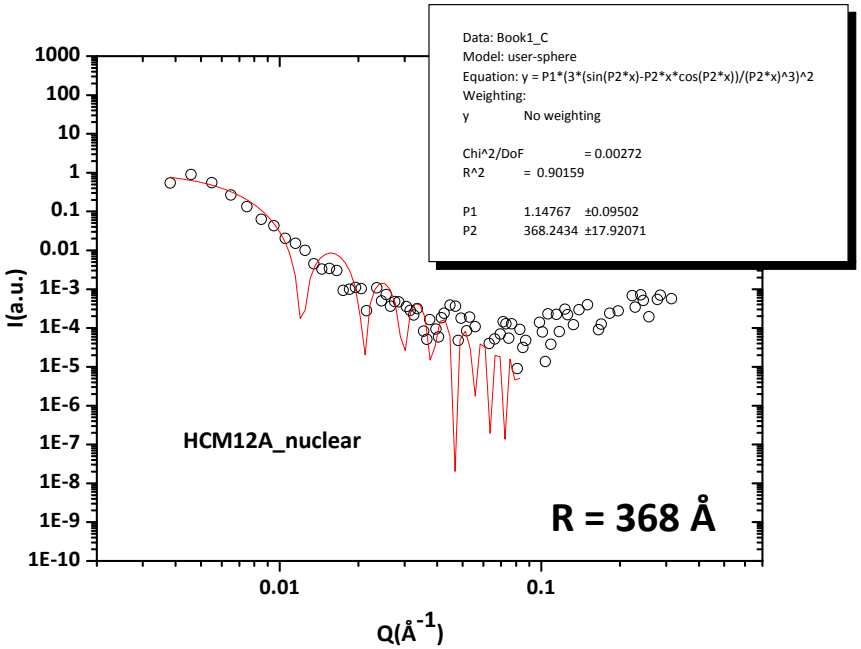

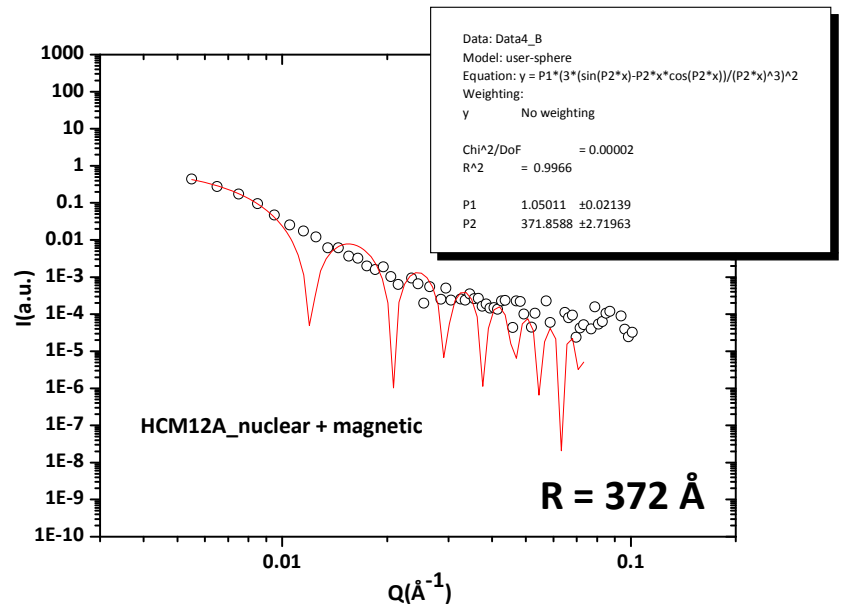

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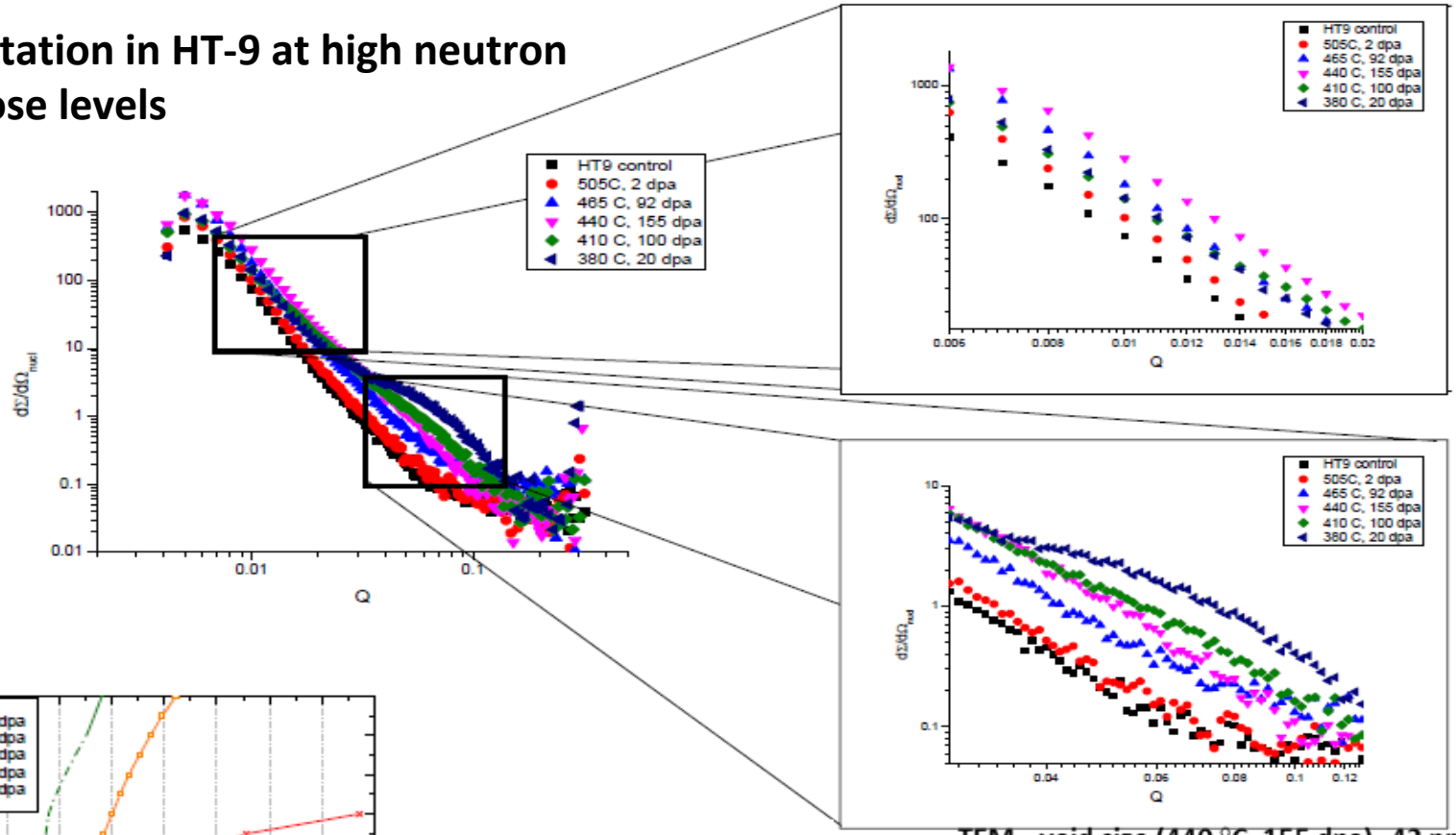
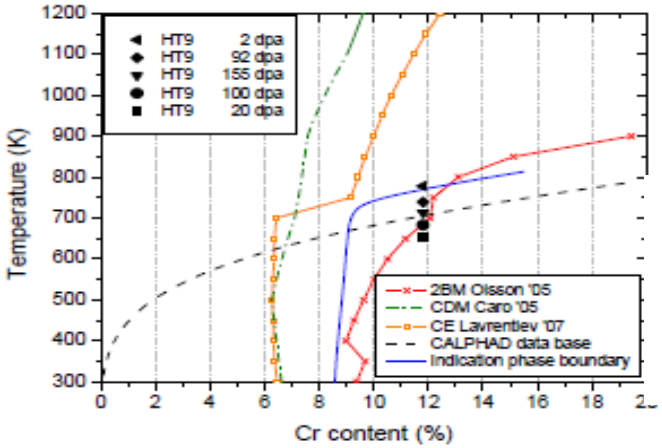
P1 nuclear + mag = 1.1 cm^{-1}
P1 control = 1.6 cm^{-1}

Phase precipitation in HT-9 at high neutron irradiation dose levels





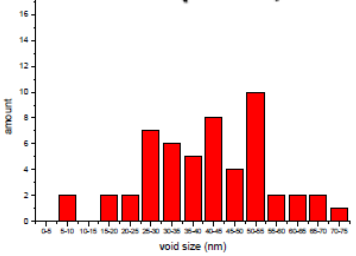
HT9

Heat 84425

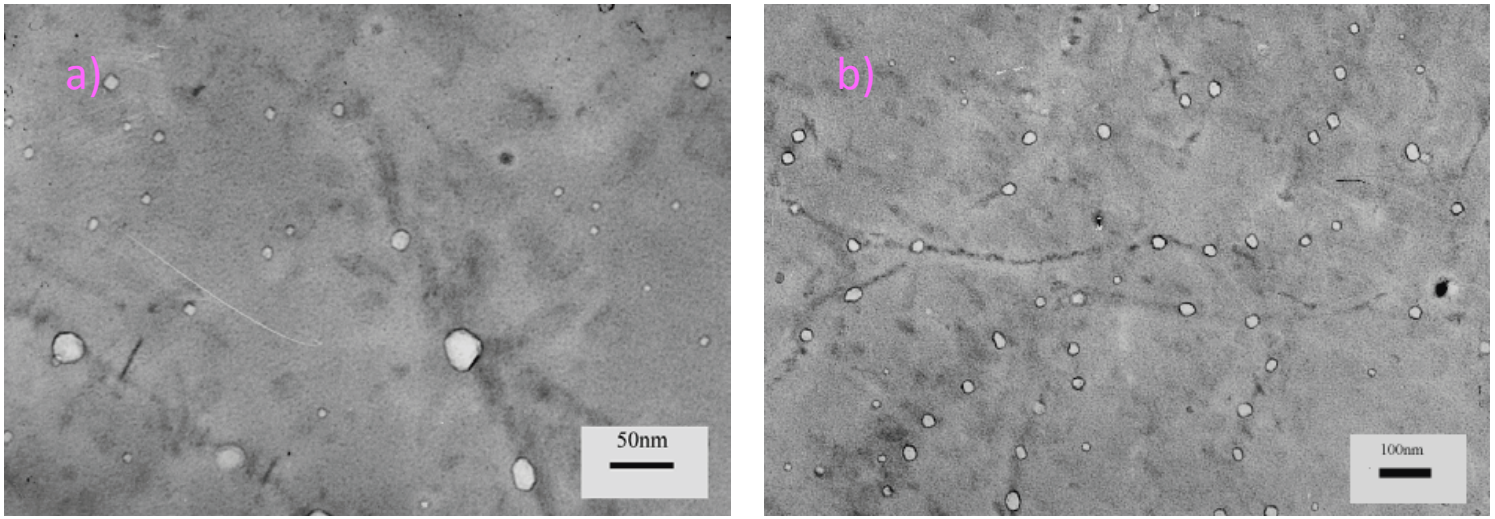
C	Cr	Mo	Mn	Si	Ni	V	W
0.21	11.8	1.03	0.50	0.21	0.51	0.33	0.52

TEM - void size (440 °C, 155 dpa), 42 nm;

TEM -void size (410 °C, 100 dpa), 23 nm;



From: F. A. Garner et al. TMS, 2005



Voids in 12X18H9T (18Cr-10Ni-Ti), analog of AISI 321 steel irradiated to 0.6 dpa at 350/430/350 °C: a) large voids on sub-grain boundaries, b) spatial distribution of smaller voids.

SANS is a complementary technique to TEM which measures similar length scales;

Differences:

- **SANS makes determinations on ensembles, TEM on specific views;**
- **SANS uses nuclear + magnetic contrast for species identification, TEM maybe limited by coherence of the crystal structures between species and matrix.**

Conclusions

- Both samples are constituted by spheres embedded in a matrix;
- Sample NF616 is constituted of spheres of an average size of 338 Å;
- Sample HCM12A is constituted of spheres of an average size of 390 Å;
- Both samples are constituted by spheres of vacancy clusters or voids;
- Models are being improved in order to provide other kind of data like the degree of polydispersity;

At LQD we've obtained information of the shape, size and type of defects in the steel matrix.