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ISIS Neutron and Muon Source

Food science research using spallation source neutrons and synchrotron source X-rays

2nd Northern Lights on Food Masterclass
15 November 2021

Gregory Smith, ISIS Neutron and Muon Source

Introduction

Colloid scientist and soft matter physicist

Currently a SANS Instrument Scientist at ISIS Neutron and Muon Source

Responsible for Larmor instrument

Previously, I worked and studied at several institutions.

- Postdoc at Niels Bohr Institute, University of Copenhagen (Denmark)
- Postdoc at Department of Chemistry, University of Sheffield (UK)
- PhD and MSci from School of Chemistry, University of Bristol (UK)

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ISIS Neutron and Muon Source

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Objectives

1

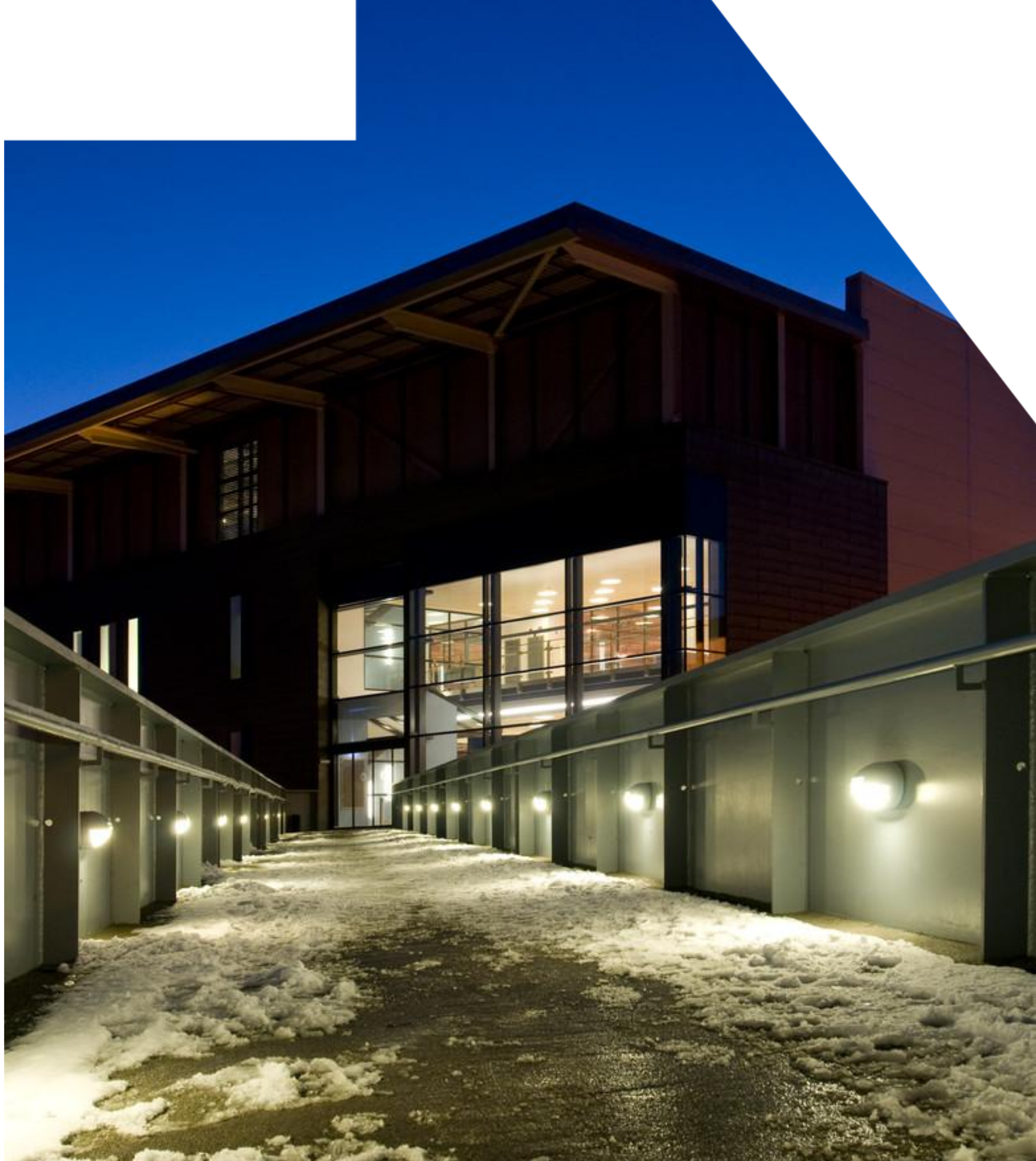
Understand how neutrons and X-rays interact differently with materials and why they are used in investigations

2

Be familiar with large-scale neutron and X-ray facilities and different types of instruments

3

Be aware of past investigations using neutrons and X-rays to study food



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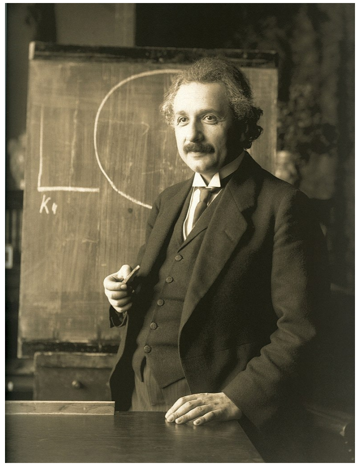
Why neutrons and X-rays?

Waves are useful tools.

Gullfoss, Iceland (Image taken by the author)

Neutrons and X-rays are waves too.

- It is not just visible light that are waves. All electromagnetic radiation (for instance, X-rays) are waves. All particles have an associated wavelength.
- Known from photoelectric effect (for light). Postulated by de Broglie for all matter.



Photons

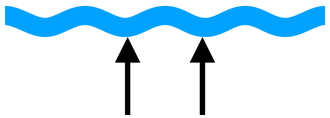
$$\lambda = \frac{h}{p}$$

Copper K-α emission wavelength is 1.54 Å

Particles

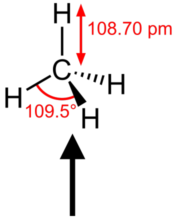
$$\lambda = \frac{h}{p} = \frac{h}{mv}$$

Free thermal neutron wavelength is ~1 Å



$\lambda_n / r_n \sim 10^6$!

Typical neutron λ (1×10^{-10} m)



C–H bond length (1×10^{-10} m)

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Albert Einstein during a lecture in Vienna in 1921. https://commons.wikimedia.org/wiki/File:Einstein_1921_by_F_Schmutzer_-_restoration.jpg
Louis de Broglie. https://commons.wikimedia.org/wiki/File:Broglie_Big.jpg

Visible light

The appearance of a camera under visible light is unsurprising and expected.

For some parts, made of metal and plastic, all we can see is the surface.

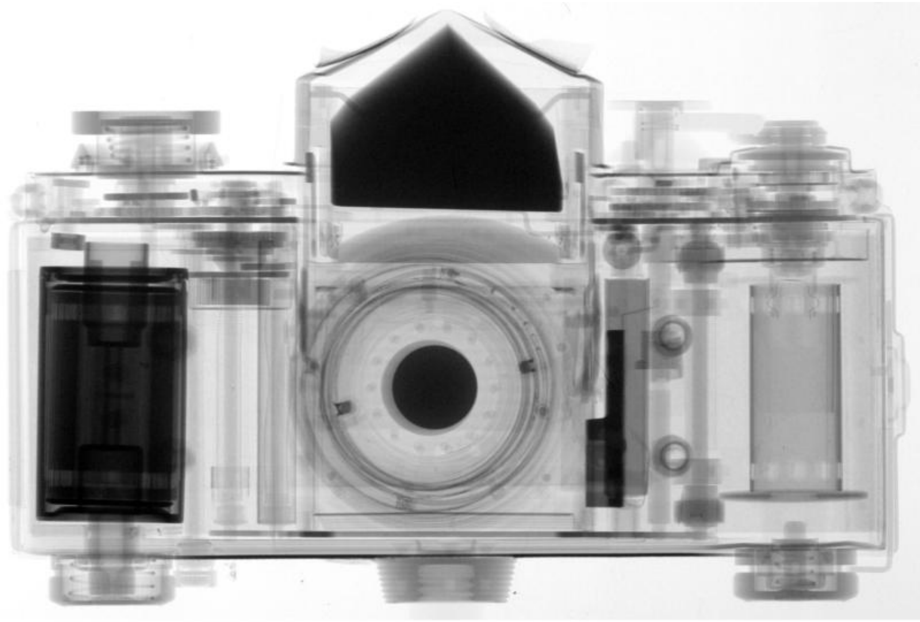
For some parts, made of glass, we can see through it as they are visible light transparent.

Maja Dumat. Canon AE-1 + FD 28mm 1:2. https://flic.kr/p/qPWnk5

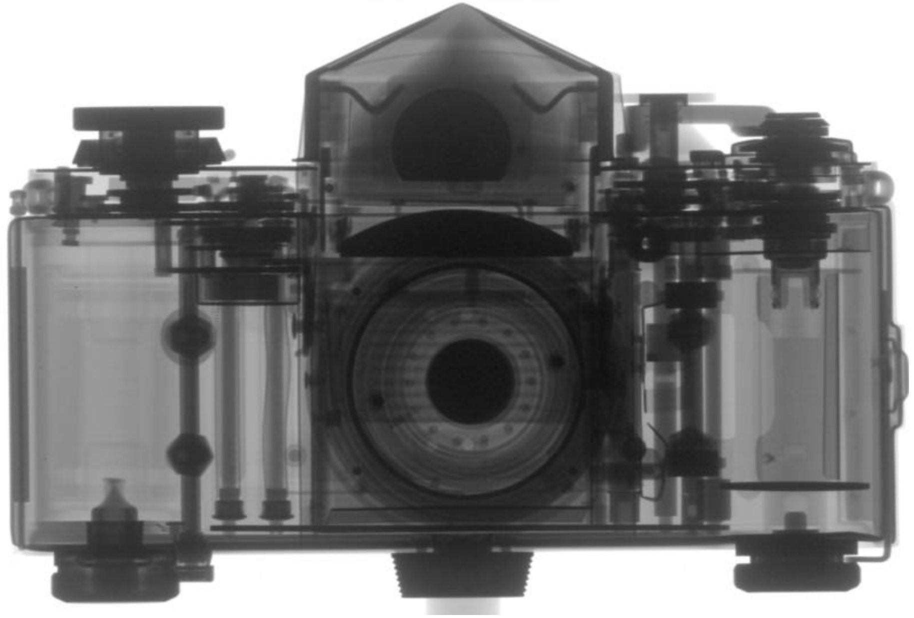
Neutrons and X-rays

The appearance of a camera imaged by X-rays and neutrons is more surprising.

The radiographs at the right show how different these types of radiation interact with the materials. X-rays are attenuated by heavier elements, such as metals. Neutrons are attenuated by hydrogen-rich compounds, such as plastic parts.



X-ray image



Neutron image

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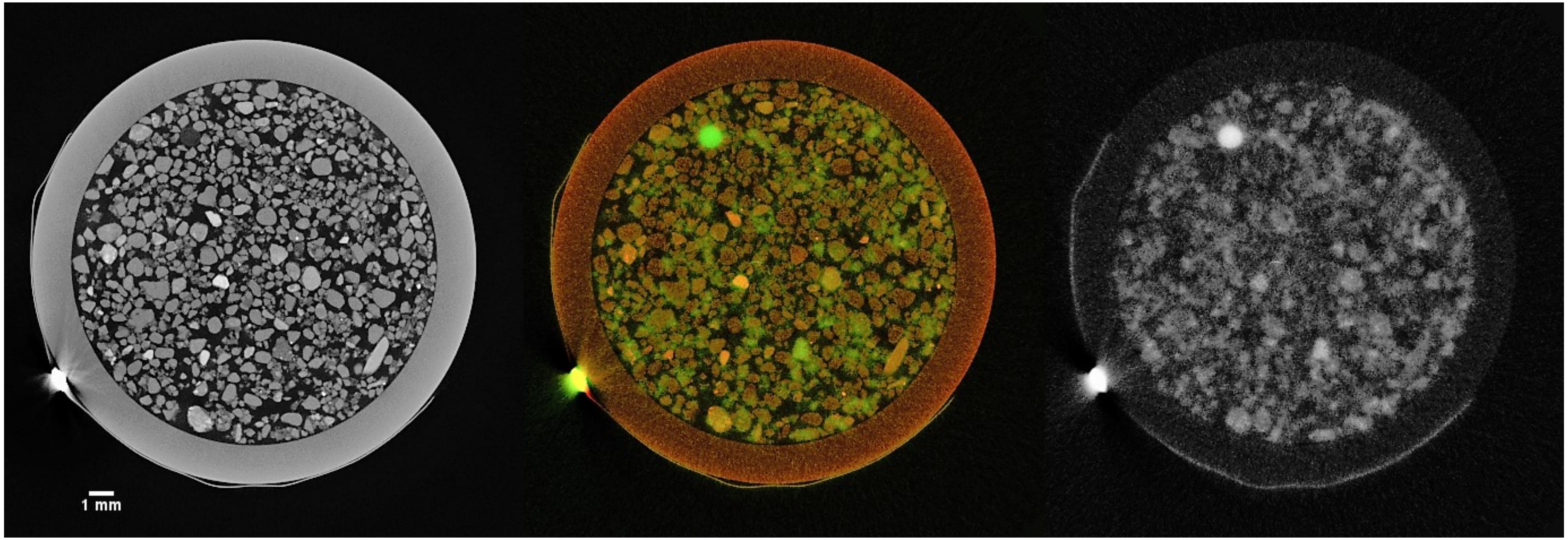
Paul Scherrer Institut. Radiograph of an analog camera: by neutrons (top) by X-rays (bottom). https://www.psi.ch/en/niag/what-is-neutron-imaging

An example closer to food (plant root)

X-ray image
Soil composition and structure



Neutron image
Water and biological matter

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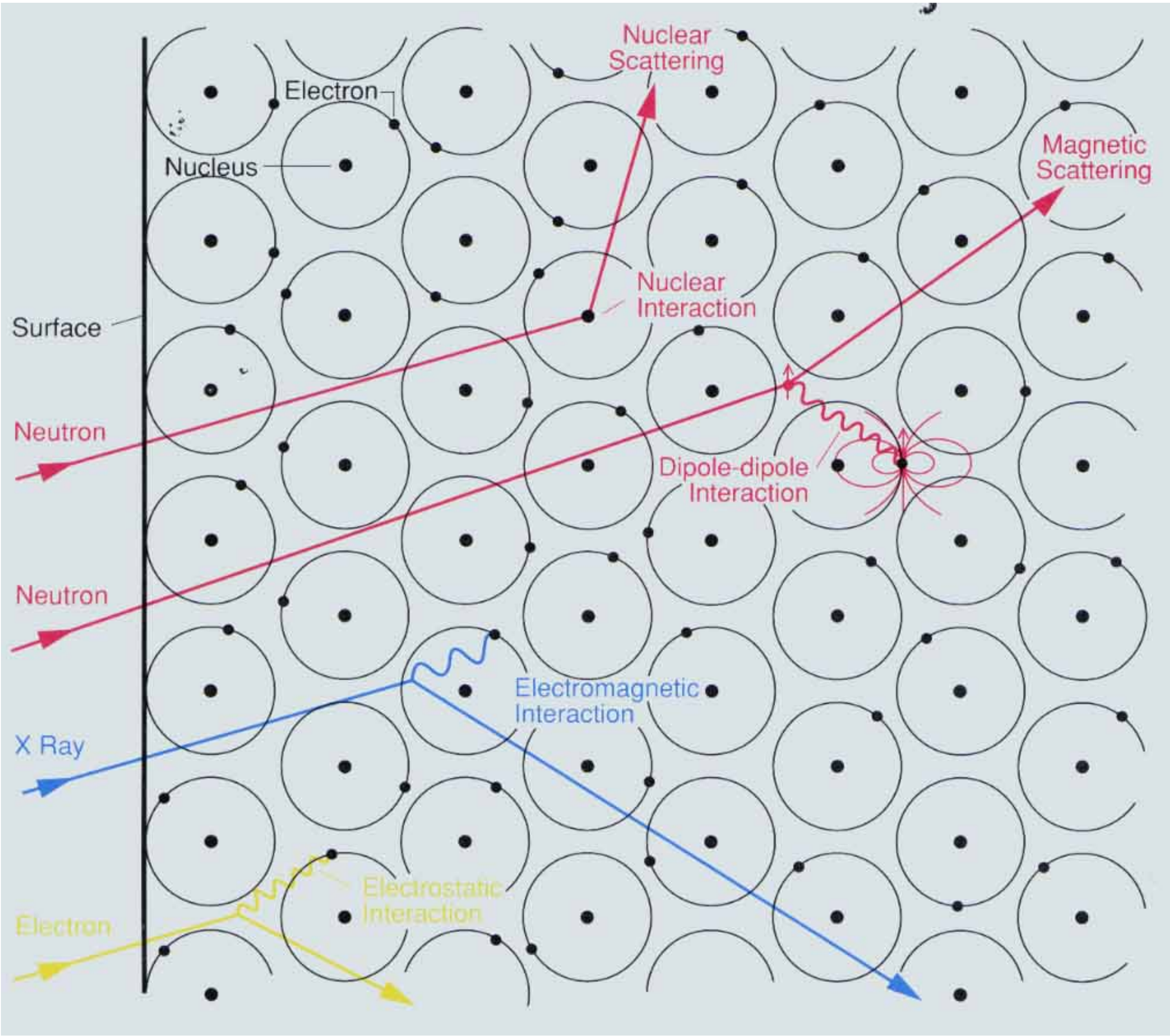
ISIS. Getting to the root of it using neutron and X-ray imaging. https://www.isis.stfc.ac.uk/Pages/SH19_RootsonIMAT.aspx
From Clark T *et al.* 2019 *J. Microsc.* **277** 170–178 doi:10.1111/jmi.12831

Interaction between radiation and matter

- Radiation interacts differently with matter.
  - X-rays interact with electrons via electromagnetic interactions.
  - Neutrons interact with nuclei via nuclear interactions (also magnetically).
- This determines two things, primarily.
  - the information that can be gained in a measurement
  - how far the radiation can penetrate



Pynn R 1990 Neutron Scattering—A Primer *Los Alamos Sci.* **19** 1–31

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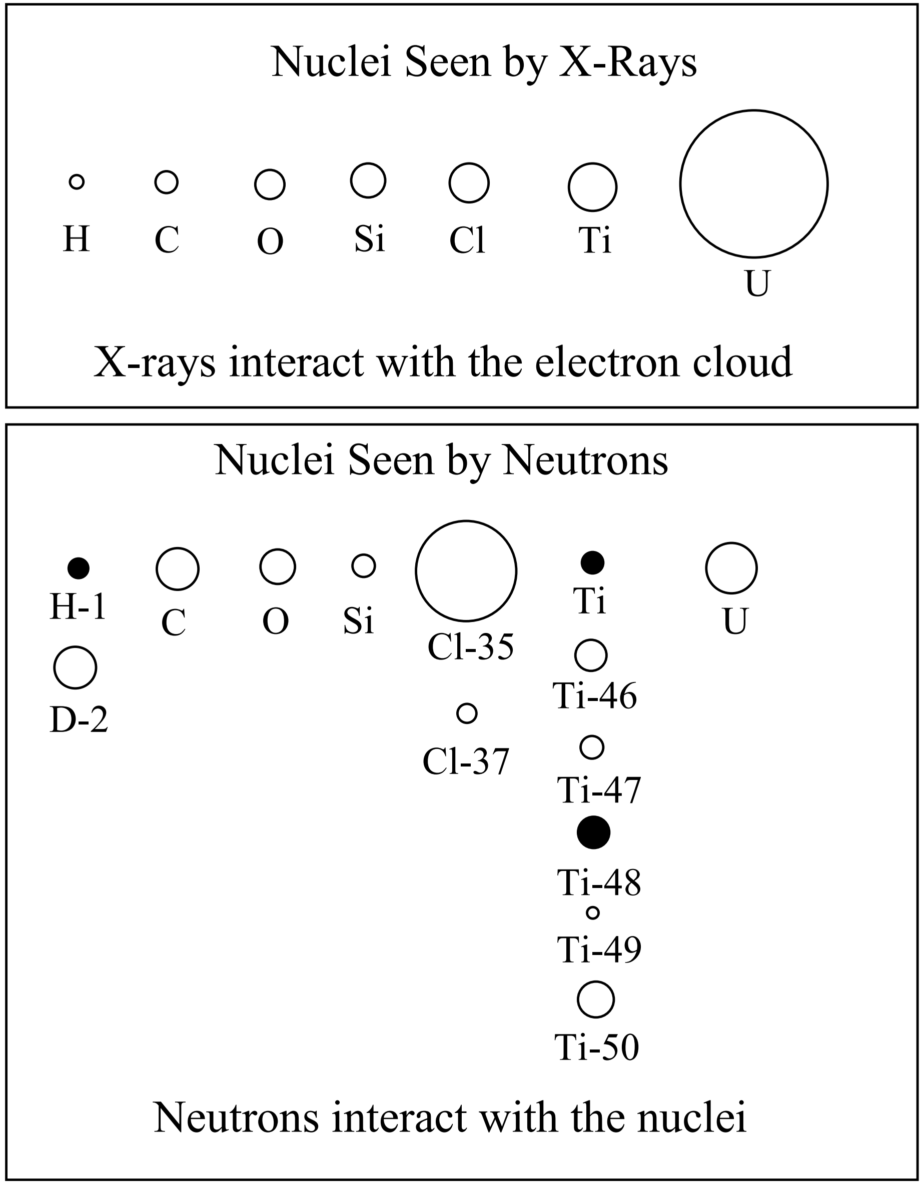
Comparing neutrons and X-rays

- Neutron and X-ray techniques are complementary.
- X-ray interactions are stronger with large *Z* elements. Neutron interactions vary erratically.



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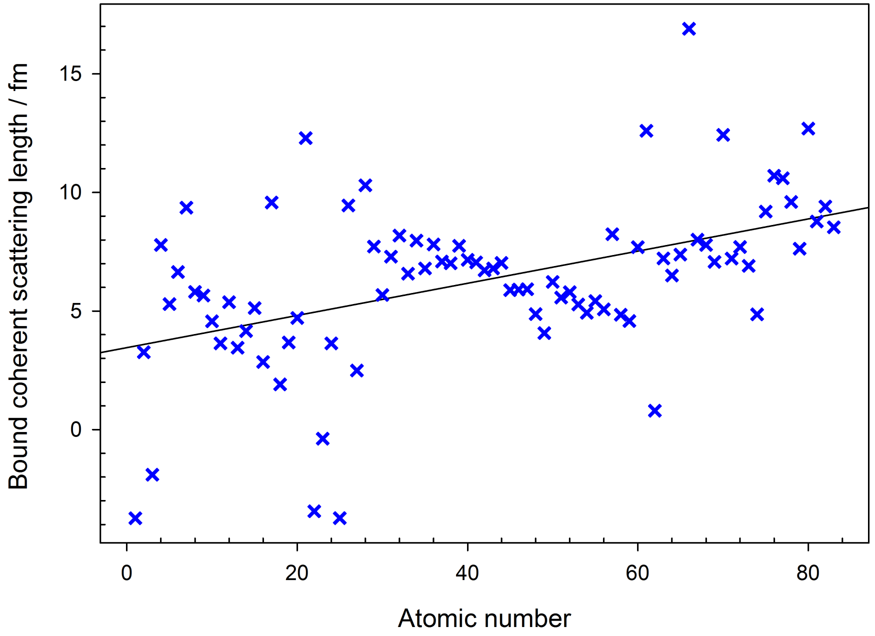
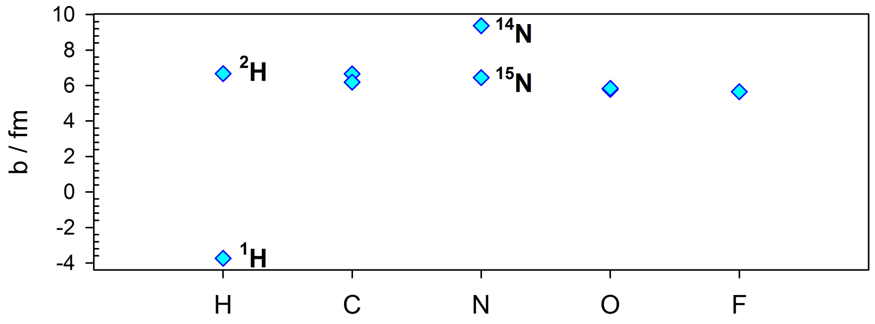
Hammouda B Probing Nanoscale Structures – The SANS Toolbox http://www.ncnr.nist.gov/staff/hammouda/the_SANS_toolbox.pdf

Comparing neutrons and X-rays

- Neutron and X-ray scattering are complementary.
- X-ray interactions are stronger with large *Z* elements. Neutron interactions vary erratically.
- Neutrons interact different with different isotopes (crucially ^{1}H vs ^{2}H).



(a) Scattering lengths of all elements from H to Np.



(b) Scattering lengths of selected small atomic number nuclei.

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Smith G N 2015 Charging colloids in nonpolar solvents PhD Thesis University of Bristol EThOS ID: uk.bl.ethos.683919
Values from Sears V F 1992 *Neutron News* **3** 26–37 doi:10.1080/10448639208218770

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Neutron and X-ray facilities

ESS and MAX IV

Science Village in Lund will host both MAX IV and the ESS. Lund University will move some activities there, and the site will host other research and innovation activities.

Vetenskapsrådet. National collaboration to maximise the benefit to Sweden of ESS and MAX IV. https://www.vr.se/images/18.1af93abe17437c1d3f4f6e/1600947513919/ESS_BIG_VIEW.jpg

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ISIS and Diamond

The Rutherford Appleton Laboratory near Oxford hosts both the UK's neutron source (ISIS) and X-ray source (Diamond). The site also hosts the Central Laser Facility (CLF) as well as space and scientific computing groups.

Science and Technology Facilities Council. Aerial view. https://www.flickr.com/photos/stfcpix/16889192337/

ISIS Neutron and Muon Source

ISIS Overview Animation with VO & Subtitles. https://vimeo.com/7813721

Diamond Light Source

diamond

What is Diamond Light Source? https://youtu.be/1kJV78_I09w

What are the advantages?

- For both spallation neutron sources and synchrotron X-ray sources, I think the main advantage is the ability to **resolve and control the energy**.
  - For spallation source neutrons, this is the ability to perform neutron time-of-flight detection.
  - For synchrotron X-ray sources, this is the ability to select wavelengths or energies for an instrument using insertion devices.

Neutron time-of flight

- Neutrons move at a velocity less than light. The time (t) between a pulse and a neutron detection event is recorded, and the neutrons travel a known distance (l). This gives the velocity (v).

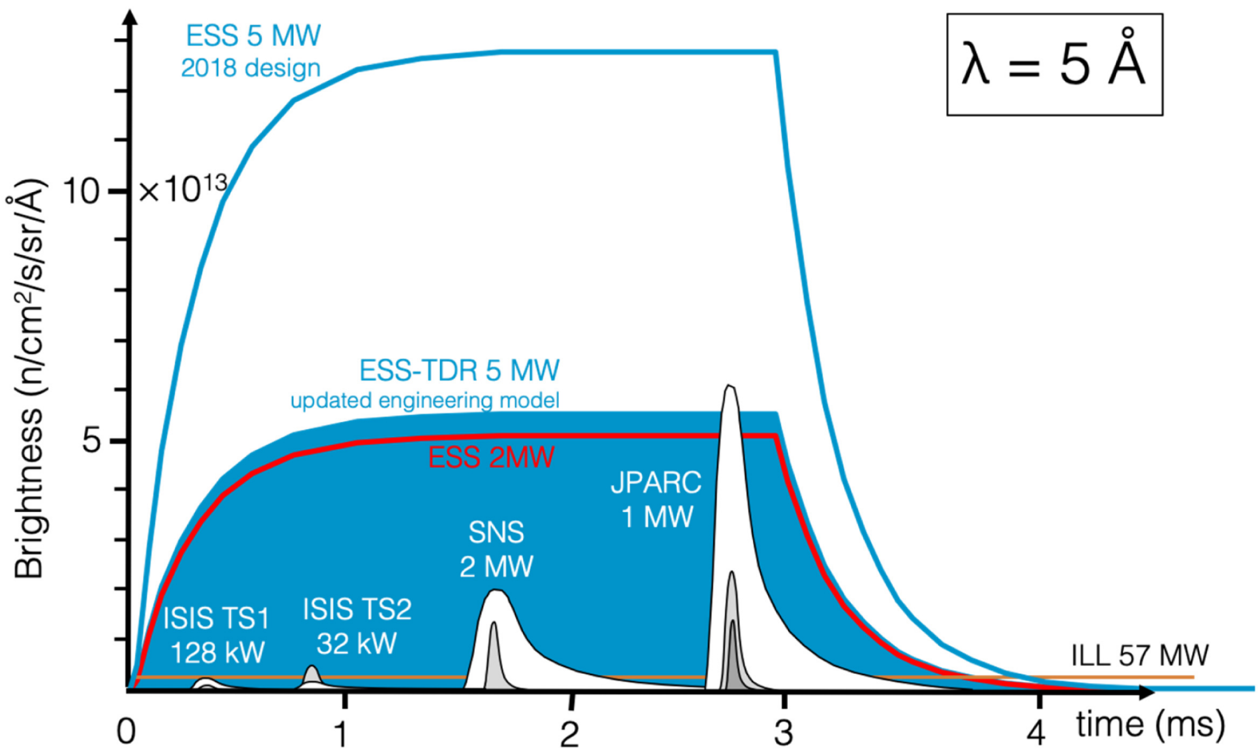
$$v = l/t$$

- From this, the kinetic energy (E_k) of the neutron can be calculated.

$$E_k = 1/2 \cdot mv^2$$

- Using de Broglie relationship and either the kinetic energy (E_k) or the momentum (p), the neutron wavelength can be calculated.

Time-dependent brightness of the ESS cold moderator



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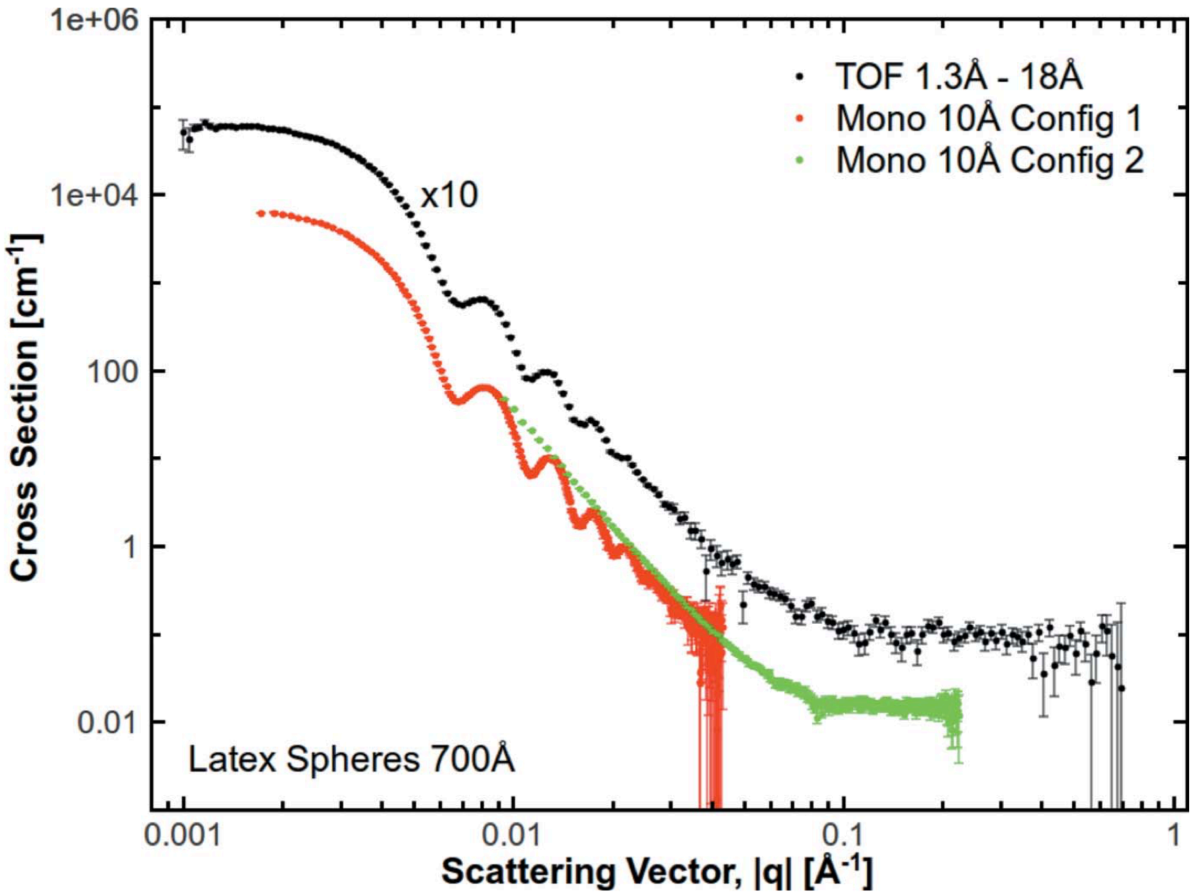
Andersen K H *et al.* 2020 *Nucl. Instrum. Method Phys. Res. A* **957** 163402 doi:10.1016/j.nima.2020.163402

Neutron time-of flight

D33 at the Institut Laue–Langevin (ILL)



Why is this an advantage?

- This can be clearly seen by looking at data from a reactor instrument that can also be run in time-of-flight mode.
- The ToF measurements were made at one instrument configuration with two detectors (front and rear), and they cover a wider *Q* range than the monochromatic measurements even when two configurations were measured.
- The are two main advantages of ToF.
    - measuring a wide wavelength range simultaneously
    - event mode enabling post-measurement time slicing

Dewhurst C D *et al.* 2016 *J. Appl. Cryst.* **49** 1 doi:10.1107/S1600576715021792 (D33, ILL)

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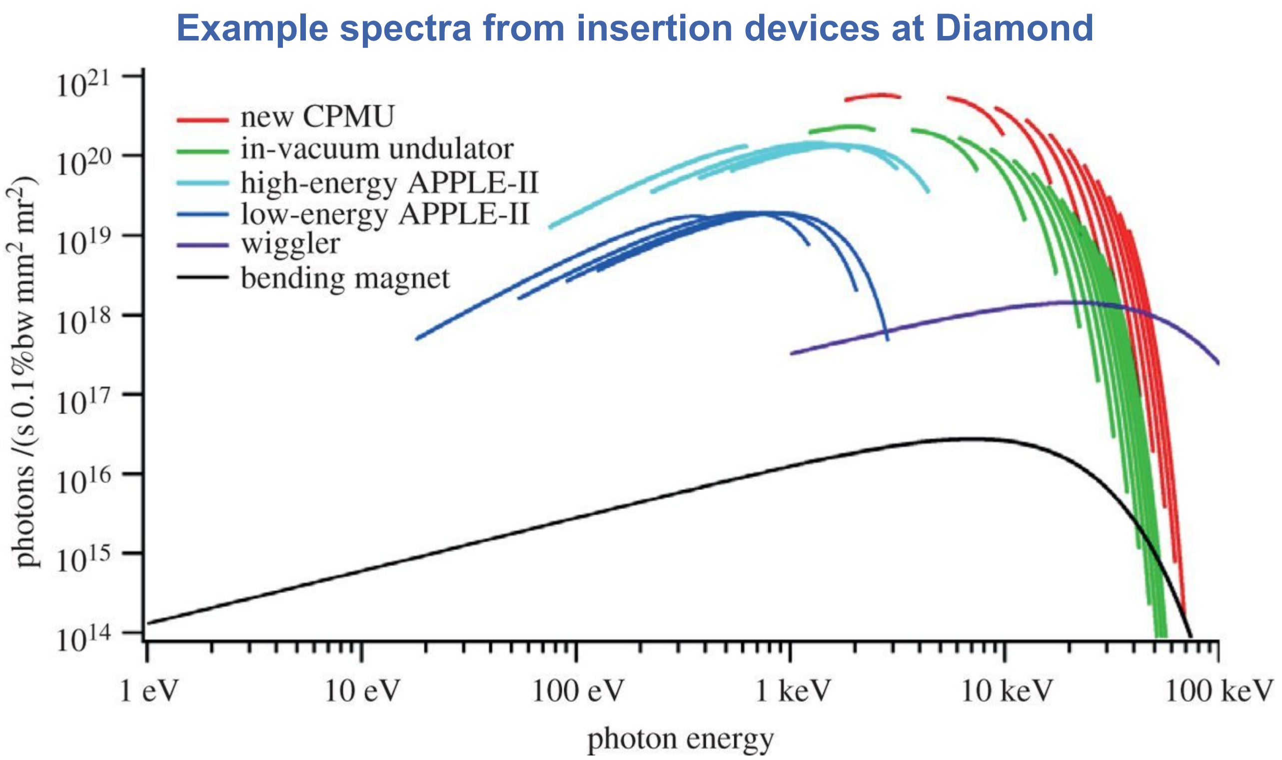
Synchrotron insertion devices

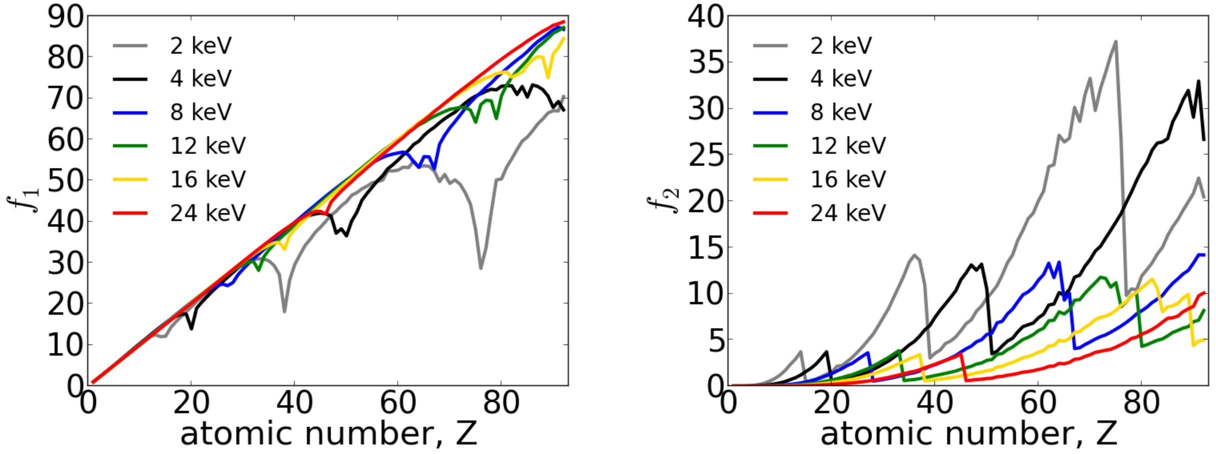


- For laboratory sources, the X-ray source is typically monochromatic with a wavelength determined by the metal anode (for instance, Cu K-α wavelength of 1.54 Å).
- Insertion devices are placed in the beam to produce brighter and more tuneable photons than possible just by bending the beam.
- At Diamond, different types of insertion devices to produce lower or higher energy photons, depending on the requirements of the beamline.

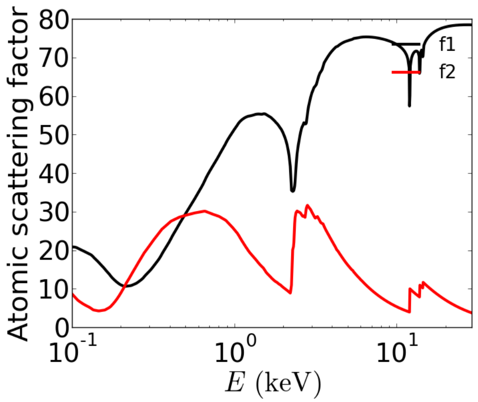
Materlik G, Rayment T, and Stuart D I 2015 *Phil. Trans. R. Soc A* **373** 20130161 doi:10.1098/rsta.2013.0161

Synchrotron insertion devices

Elemental dependence of atomic scattering factors



Energy dependence for Au



Why is this an advantage?

- X-ray atomic scattering factors are not independent of the energy of the photon. There are large and abrupt changes in either absorption (f_1) or fluorescence (f_2) near absorption edges.
- This is essential for spectroscopy measurements using synchrotrons.
- It also enables scattering measurements that are used to gain elemental contrast by tuning to absorption edges.

GISAXS. Atomic scattering factors. http://gisaxs.com/index.php/Atomic_scattering_factors

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Neutron and X-ray techniques

What kinds of instruments are there?

- There is a **huge** variety.
- There are instruments from many areas of science, over a wide range of length scales.
- Comparing neutron and X-ray sources, there are some techniques in common, and some that take advantage of the properties of neutrons or photons.

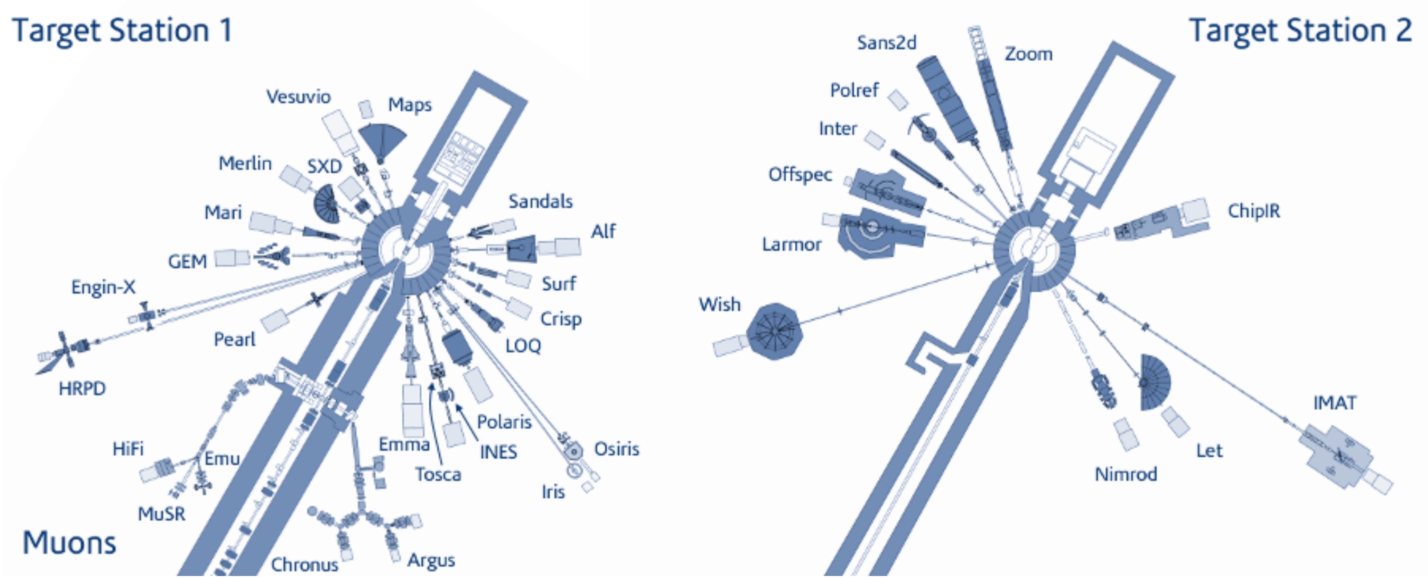
What kinds of instruments are there?

Facilities generally divide instruments into four categories

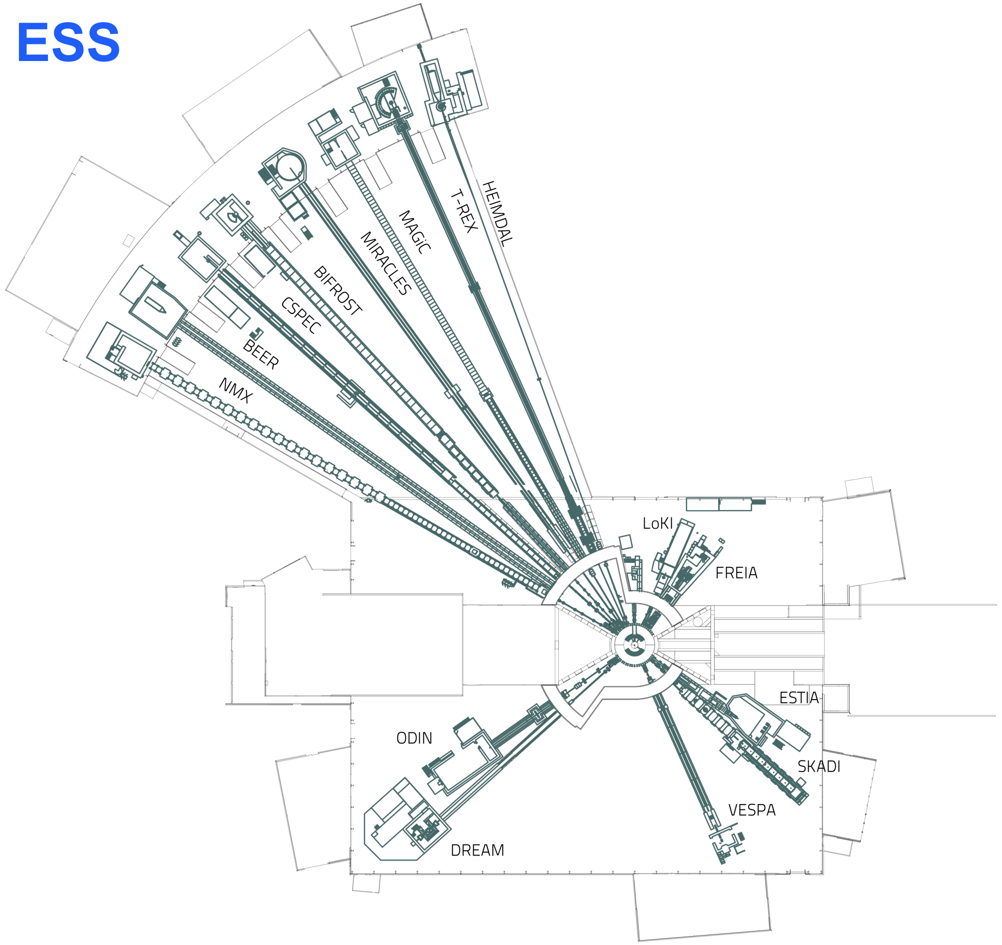
- Imaging
- Large-scale structures
- Diffraction
- Spectroscopy

ISIS and ESS

ISIS



ESS



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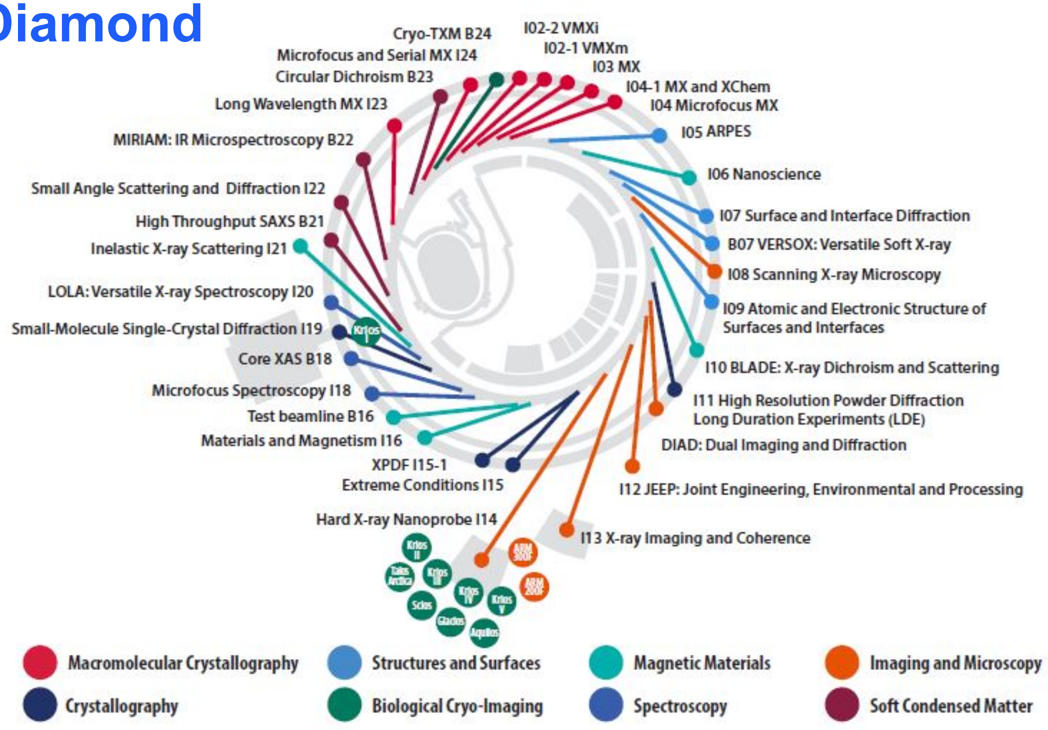
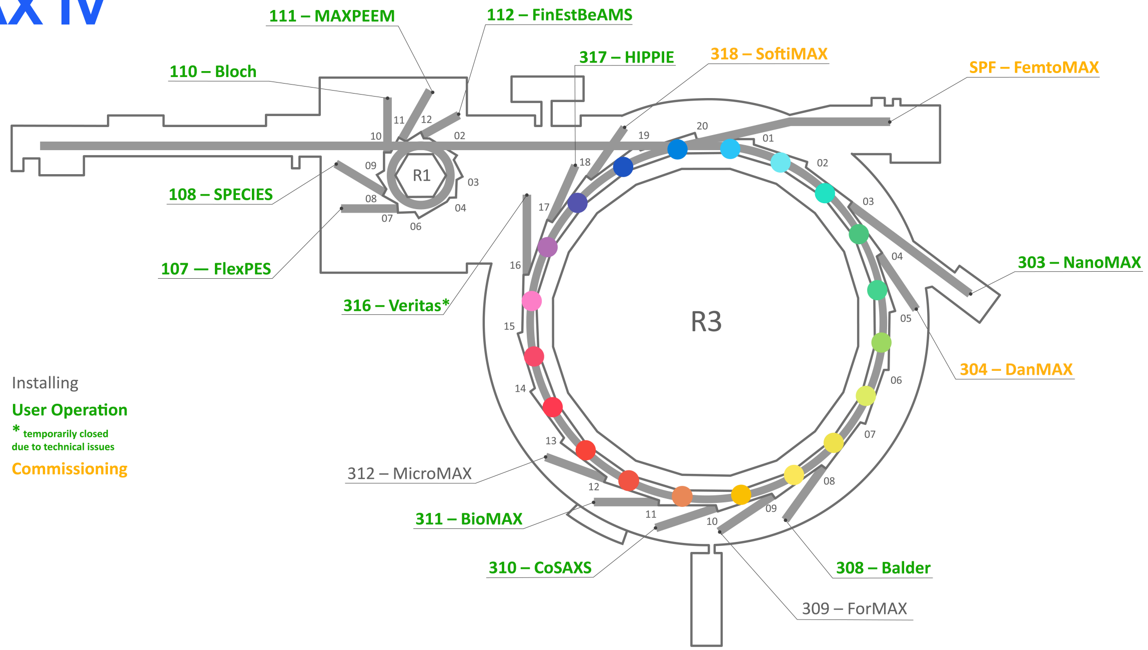
ISIS https://www.isis.stfc.ac.uk/Pages/Instruments.aspx
ESS https://europeanspallationsource.se/instruments

Diamond and MAX IV

Diamond



MAX IV



Diamond https://www.diamond.ac.uk/Home/Corporate-Literature/Annual-Review/Review2019/Beamline-Development-and-Technical-Summary.html
MAX IV https://www.maxiv.lu.se/accelerators-beamlines/beamlines/

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Neutrons and X-rays in Food Science

Thank you to…

Sarah Rogers and Christ Frost (ISIS)
Claire Pizzey (Diamond)

Literature examples

Trends in Food Science & Technology
Volume **20**, Pages 576–586
doi:10.1016/j.tifs.2009.07.008

In the following slides, I am going to discuss examples from ISIS and Diamond. These were chosen from the main instrument categories.

- Imaging
- Large-scale structures
- Diffraction
- Spectroscopy

Elliot Gilbert (ANSTO) has published several excellent reviews on neutron and scattering techniques and food science.

- Characterisation Techniques in Food Materials Science doi:10.1002/9781118373903.ch3
- Small-angle X-Ray and neutron scattering in food colloids doi:10.1016/j.cocis.2019.03.005
- Food Structure Characterisation Using Small-angle Scattering Methods doi:10.1039/9781788016155-00309

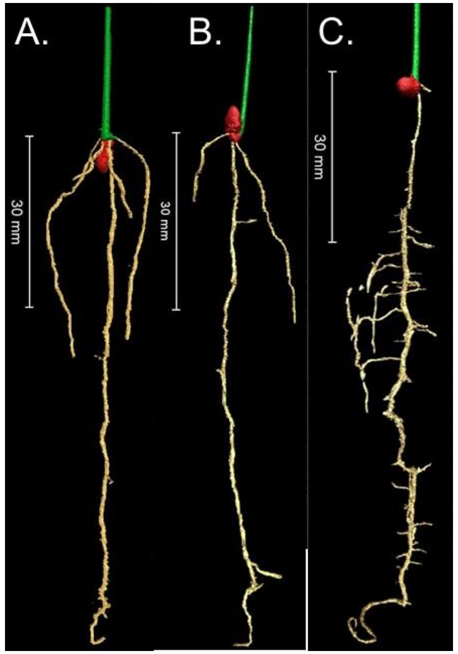
Neutron scattering: A natural tool for food science and technology research

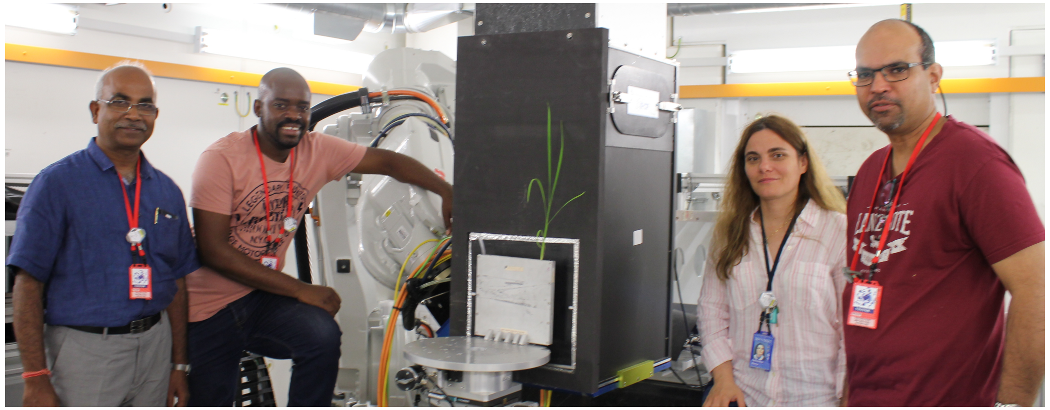
Amparo Lopez-Rubio[a] and Elliot Paul Gilbert[b,*]

[a]Novel Materials and Nanotechnology Laboratory, IATA-CSIC, Apdo. correos 73, 46100 Burjassot, Valencia, Spain
[b]Bragg Institute, Australian Nuclear Science and Technology Organisation, Private Mail Bag 1, Menai, NSW 2234, Australia (Tel.: +61 2 9717 9470; fax: +61 2 9717 3606; e-mail: elliot.gilbert@ansto.gov.au)

A greenhouse at a neutron facility

- Transporting samples to do neutron imaging can damage samples or reduce certainty about conditions.
- IMAT beamline commissioned the first research greenhouse at a neutron facility.
- The research greenhouse makes preparation of plant samples as easy as using a chemical preparatory lab.
- Computerized neutron tomography enabled root system architecture to be imaged.





The University Of Sheffield.

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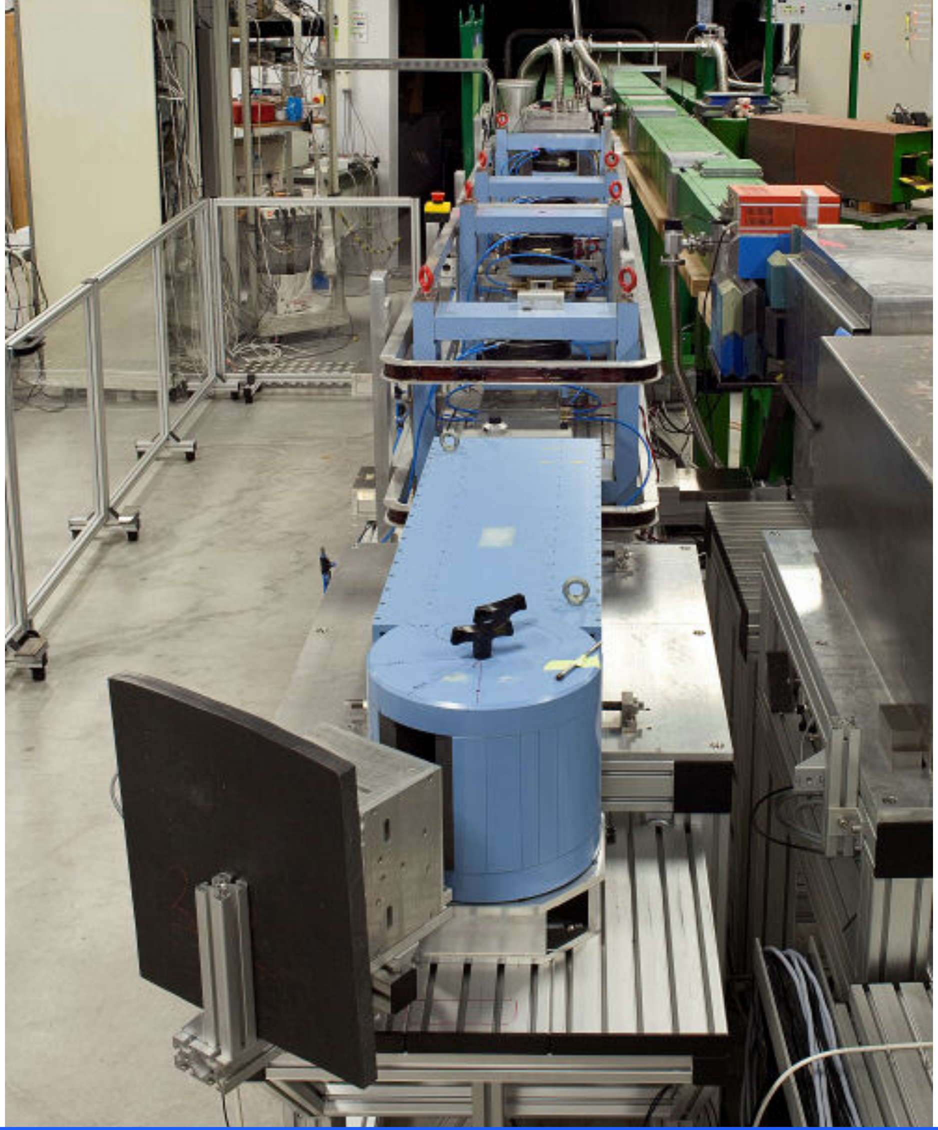
Cereal research takes roots on IMAT. https://www.isis.stfc.ac.uk/Pages/SH19_Cereal-research-on-IMAT.aspx

Mawodza T 2019 Plant-soil interactions: the impact of plant water use efficiency on root architecture and soil structure PhD Thesis Universtiy of Sheffield EThOS ID: uk.bl.ethos.798124

Yoghurt formation studied by SESANS

- SESANS stands for spin-echo SANS, a variant of SANS that can study μm length scales.
- These measurements were not actually performed at ISIS. They were actually measured at the SESANS instrument at Reactor Institute Delft at TU Delft.
- However, the Larmor instrument at ISIS also has a SESANS mode. Unlike the Delft instrument, it is a time-of-flight instrument.



Bouwman W G 2021 Spin-echo small-angle neutron scattering for multiscale structure analysis of food materials *Food Structure* **30** 100235 doi:10.1016/j.foostr.2021.100235

SESANS (TU Delft). https://www.tudelft.nl/en/faculty-of-applied-sciences/business/facilities/reactor-institute-delft/research-instruments/sesans

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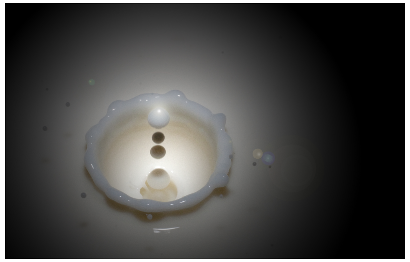
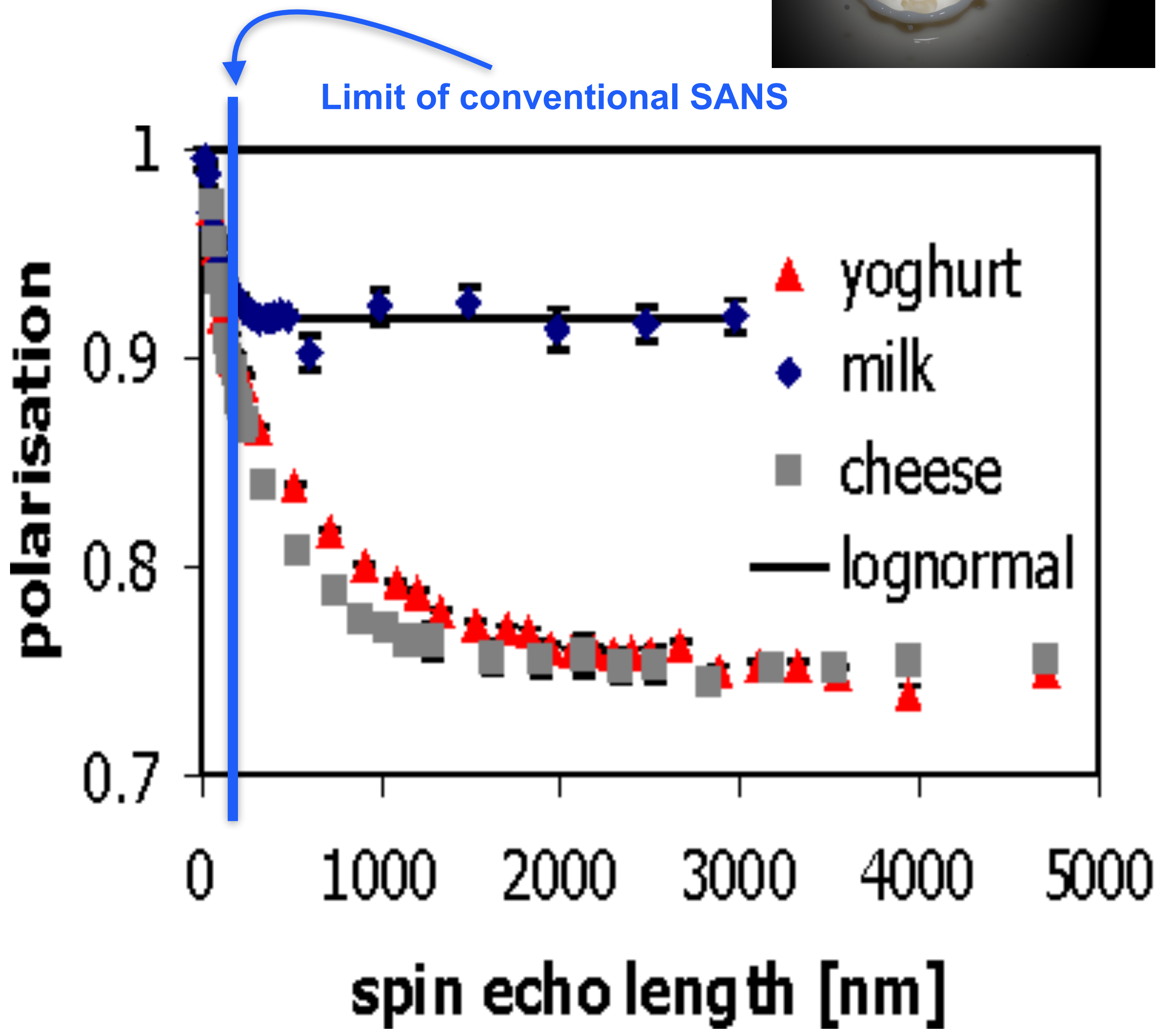
Yoghurt formation studied by SESANS



- Casein micelles are the main structural component of milk. They have a size on order 100 nm.
- “Deuterated milk” prepared by dissolving milk powder in D_2O at correct concentration.
- SESANS data on milk sample shows scattering from casein micelles only with correct size.



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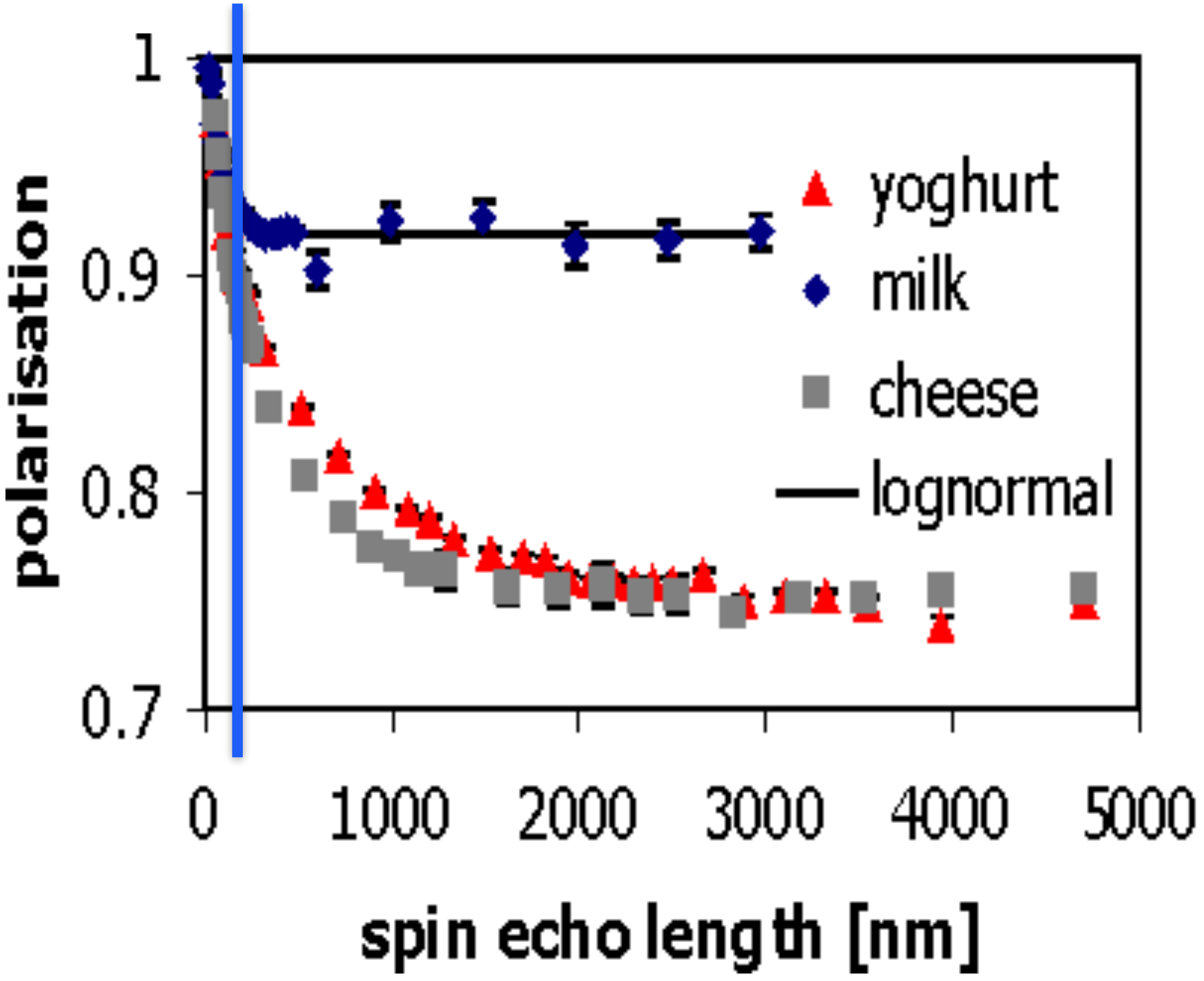
Bouwman W. Yoghurt formation studied by SESANS. http://wimbouwman.nl/dairy.php
Milk Droplet_6626 (Brian Rogers) https://flic.kr/p/7Hpczk

Yoghurt formation studied by SESANS

- Deuterated yoghurt and cheese prepared by acidifying milk (yoghurt) or adding an enzyme (cheese). Care must be taken due to D_2O isotope effects.
- SESANS data shows changes over much longer length scales (up to 3 μm) than milk. There is a difference in structure between yoghurt and cheese.



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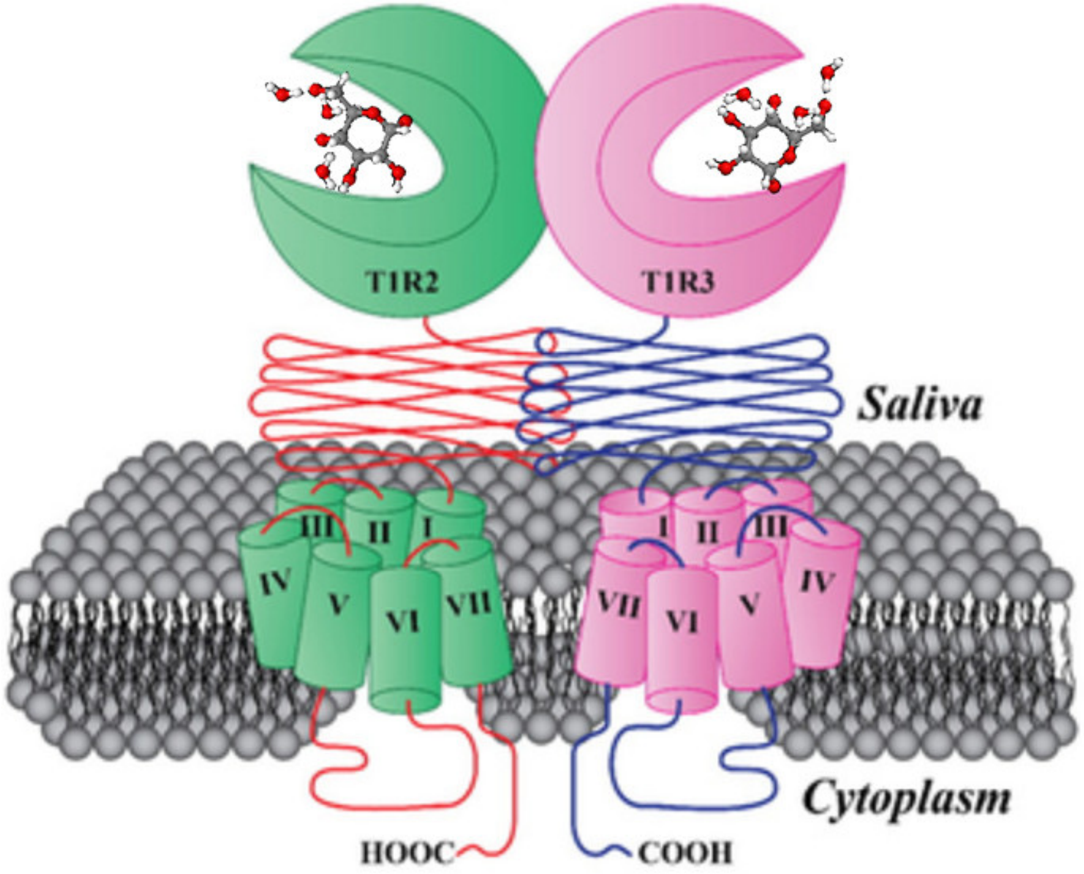
Bouwman W. Yoghurt formation studied by SESANS. http://wimbouwman.nl/dairy.php
Cheese (Niklas Morberg) https://flic.kr/p/6mCXUi, Greek Maple Sheep Milk Yogurt & Fresh Picked Raspberries (Jo Zimny says: The Garden Is Waking Up!) https://flic.kr/p/6pssZc

Linking hydration and sweetness

- The interaction between molecules and taste receptor proteins dictates how food tastes. In the case of this study, how sweet or bitter.
- Two different sugars with different degrees of perceived sweetness and bitterness were studied.
- Both are proton-rich, which is good for neutron experiments and deuteration.



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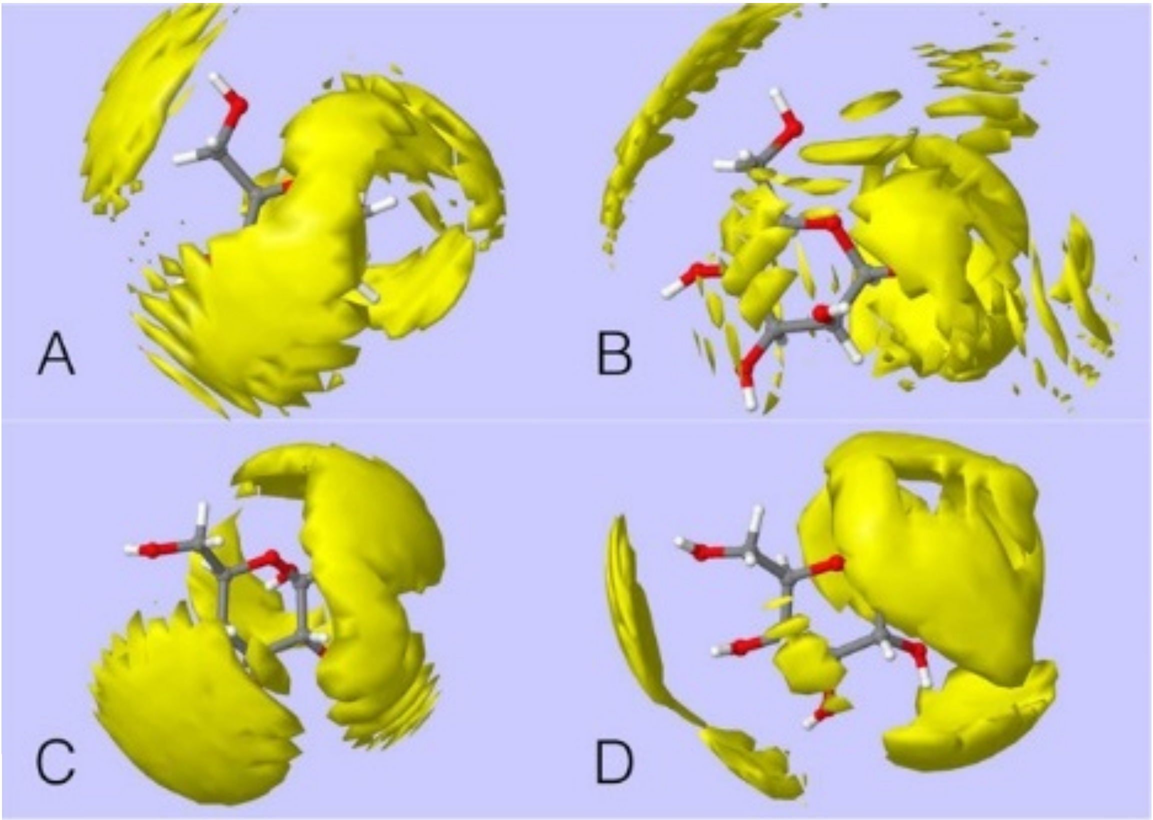
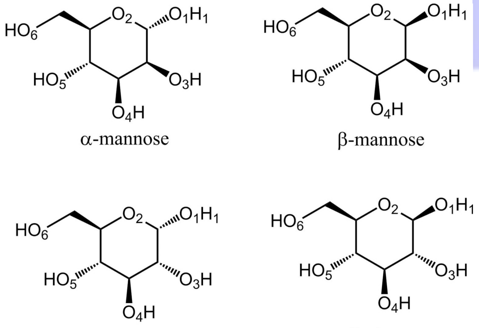
Neutrons reveal secret to sweetness. https://www.isis.stfc.ac.uk/Pages/Neutrons-reveal-secret-to-sweetness.aspx

Rhys N H *et al.* 2017 *J. Phys. Chem. B* **121** 7771 doi:10.1021/acs.jpcb.7b03919

Linking hydration and sweetness

- Diffraction measurements performed on stereoisomers of glucose and mannose.
- The extent of hydrogen bonding of the two sugars in water differs. These results were compared to models of receptor interaction.
- The greater the degree of hydrogen bonding with water, the greater the perceived sweetness of the sugar.





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Neutrons reveal secret to sweetness. https://www.isis.stfc.ac.uk/Pages/Neutrons-reveal-secret-to-sweetness.aspx
Rhys N H *et al.* 2017 *J. Phys. Chem. B* **121** 7771 doi:10.1021/acs.jpcb.7b03919

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One more example…

The structure of Limoncello

Neutrons unlock the secrets of limoncello

NEUTRONS FOR SOCIETY

If you're looking for a liquid to study with neutron scattering, lemon liqueur probably isn't your first thought. But, for Dr. Leonardo Chiappisi, a researcher at the Institut Laue-Langevin (ILL, France) and the Technical University of Berlin (TU Berlin), it was an obvious choice. "I'm originally from Sicily," he jokes, "and limoncello is the best thing to help liven up a dinner."

Limoncello, a cloudy neon-yellow liqueur, is common across southern Italy. People often make it themselves by leaving lemon zest to mature in alcohol for several weeks. It's also a personal favourite of Dr. Chiappisi and the inspiration for a paper that he hopes will help develop new speciality chemicals – especially eco-friendly products using citrus oils.

Limoncello is among a group of liqueurs that experience the so-called "ouzo effect", named after ouzo, an anise-flavoured aperitif. Ouzo is normally a clear liqueur, but it turns milky and opaque when water is added, and can stay like that for a long time.

c&en

Looking into limoncello

Credit: Laura Howes/C&EN

Mellow yellow: Limoncello, best served chilled, cloudy, and with an appreciation of the nanoscale structure within.

Running fun experiments in a scientist's spare time has a long and illustrious history, but not all researchers have access to a neutron beam source. Someone who does is chemist Leonardo Chiappisi, a Sicilian and lover of limoncello who works at the Institut Laue-Langevin, in Grenoble, France. What, he wondered, could neutron scattering tell him about his beloved aperitif?

Limoncello muss milchig sein – aber warum ist er das?

Dienstag, 18. Dezember 2018

Medieninformation Nr. 263/2018

Europäische Forschung mit Geschmack: Ein PostDoc der TU Berlin untersucht italienischen Likör in Frankreich

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Nach der Weihnachtsgans, der französischen Foie Gras oder dem italienischen Festtags-Menü schmeckt er besonders gut: Ein eiskalter Limoncello als flüssiger Nachtisch. Der süße, milchig-neongelbe, italienische Likör ist europaweit nicht nur ein beliebter Digestif oder Basis für originelle Cocktails, sondern auch ein Objekt wissenschaftlicher Neugierde: „Uns interessiert vor allem der sogenannte ‚Ouzo-Effekt' des Limoncello", so Dr. Leonardo Chiappisi, PostDoc an der TU Berlin und dem Institut Laue-Langevin (ILL, Frankreich), wo die stärkste Neutronenquelle der Welt beheimatet ist. „Benannt ist dieser Effekt nach Ouzo, einem Anis-Aperitif, bei dem es sich normalerweise um eine klare Flüssigkeit handelt, die durch Zugabe von Wasser milchig und undurchsichtig wird. Uns interessierten die physikalischen Grundlagen für diese stabile Mischung aus Alkohol, Öl und Wasser", so der geborene Sizilianer, der gerne bekennt, dass seine Herkunft die Auswahl des Untersuchungsobjektes nicht unwesentlich beeinflusst hat. „Limoncello ist in ganz Süditalien verbreitet. Viele Italiener stellen den Zitronenlikör selbst her,

le dauphiné libéré

› Edition Grenoble / Vercors › Grenoble

Grenoble - Grâce aux neutrons, deux chercheurs dévoilent (en partie) les secrets de la célèbre liqueur italienne

Le limoncello comme vous ne l'avez jamais vu

Deux chercheurs de l'Institut Laue-Langevin de Grenoble ont mis au jour les secrets de la structure du limoncello, grâce aux neutrons. Leurs résultats ouvrent un nouveau champ de recherche scientifique.

Par **Séverine MERMILLIOD** - 27 mars 2019 à 06:00 | mis à jour le 27 mars 2019 à 12:37 - Temps de lecture : 3 min

Vu 93 fois

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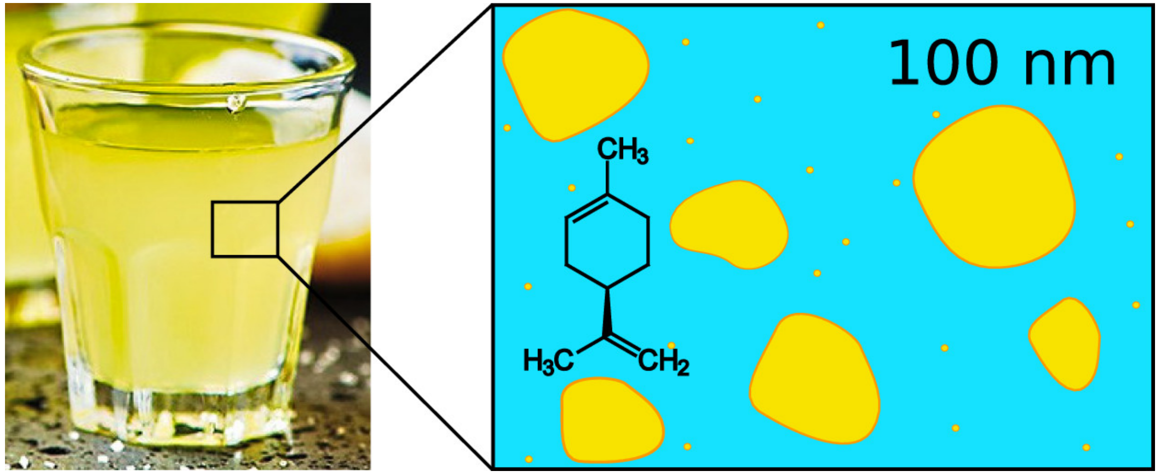
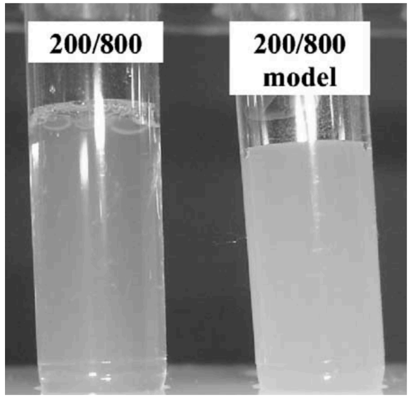
Looking into Limoncello



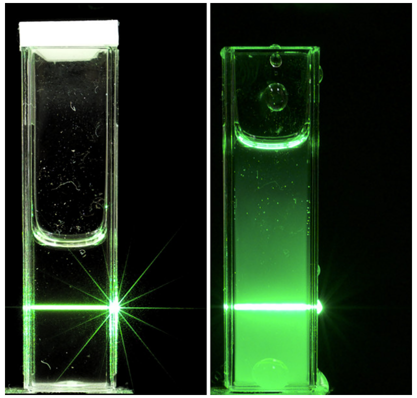
Why do I like this example so much?

- There are already examples of the physics of drinks such as pastis or absinthe in the literature.
- What I like about this study into Limoncello is the amount of work that the authors did to prepare a deuterated yet realistic drink for their neutron scattering measurements.

Pastis (2003)



Absinthe (2021)



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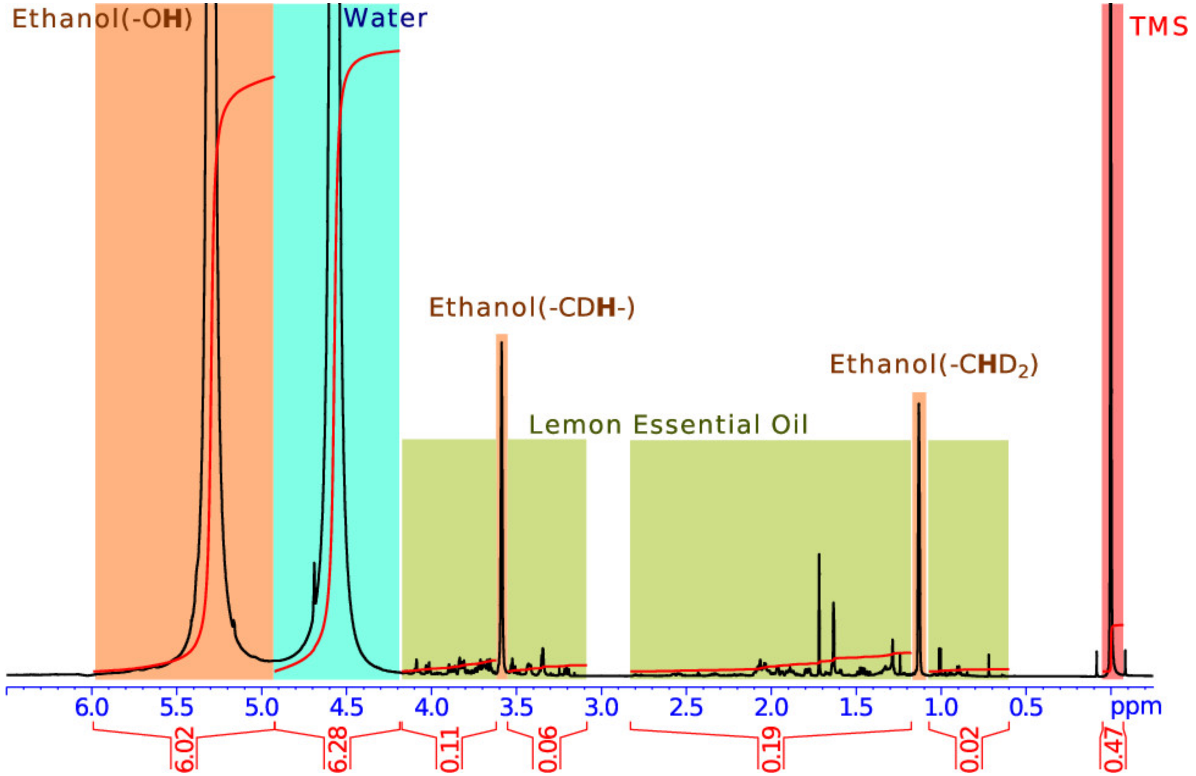
Chiappisi L and Grillo I 2018 *ACS Omega* **3** 15407 doi:10.1021/acsomega.8b01858
Grillo I 2003 *Colloids Surf. A* **225** 153 doi:10.1016/S0927-7757(03)00331-5 (Pastis)
Bickel J E *et al.* 2021 *ACS Omega* **6** 17674 doi:10.1021/acsomega.1c02246 (Absinthe)

Preparing Limoncello

- Limoncello is clearly more than just a water-ethanol mixture. The two fluids are entirely miscible. Limoncello is not homogenous.
- The traditional Limoncello recipe involves macerating lemon peel in 95% ethanol for several weeks. This extracts essential oils, which the authors identify using ^{1}H NMR.



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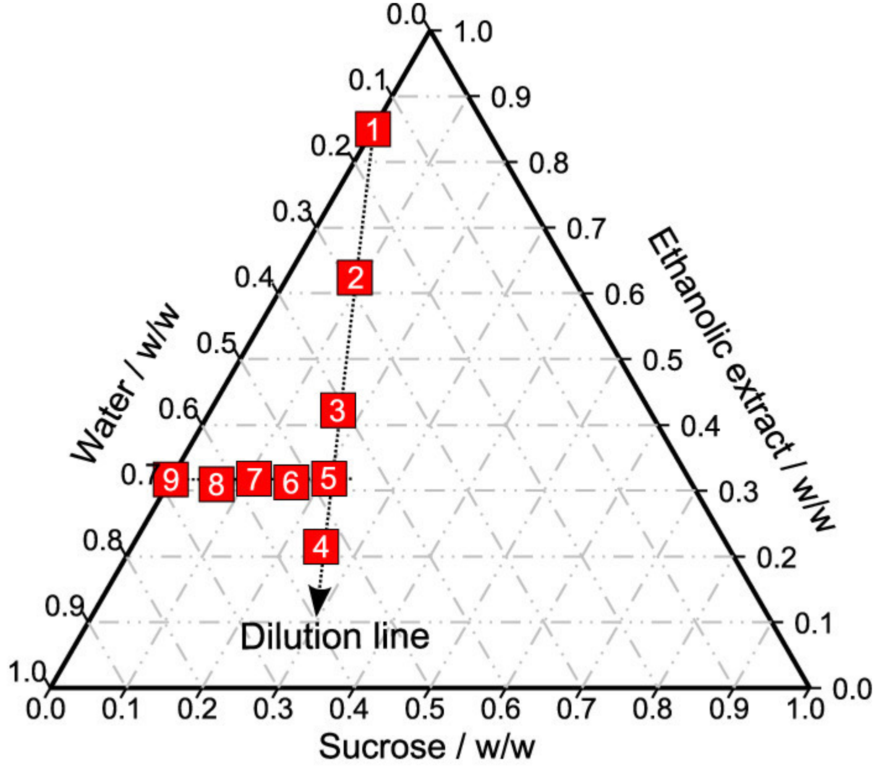
Preparing Limoncello

The samples were prepared using deuterated water, d_6-ethanol, commercial sucrose, and nontreated lemons from Sicily bought at a local market. The ethanolic extract was prepared by immersing 2.5 g of the outer part of lemon skins, the flavedo, in 12.6 g of **pure fully deuterated ethanol**. The lemon peels were kept macerating in the absolute deuterated alcohol for 4 weeks. After this period, a **sucrose syrup** was prepared by dissolving 6.6 g of sucrose in 13.2 g of **D_2O**. Samples were prepared by addition of the syrup and/or D2O to the ethanolic extract. Gentle shaking is sufficient for homogenization.



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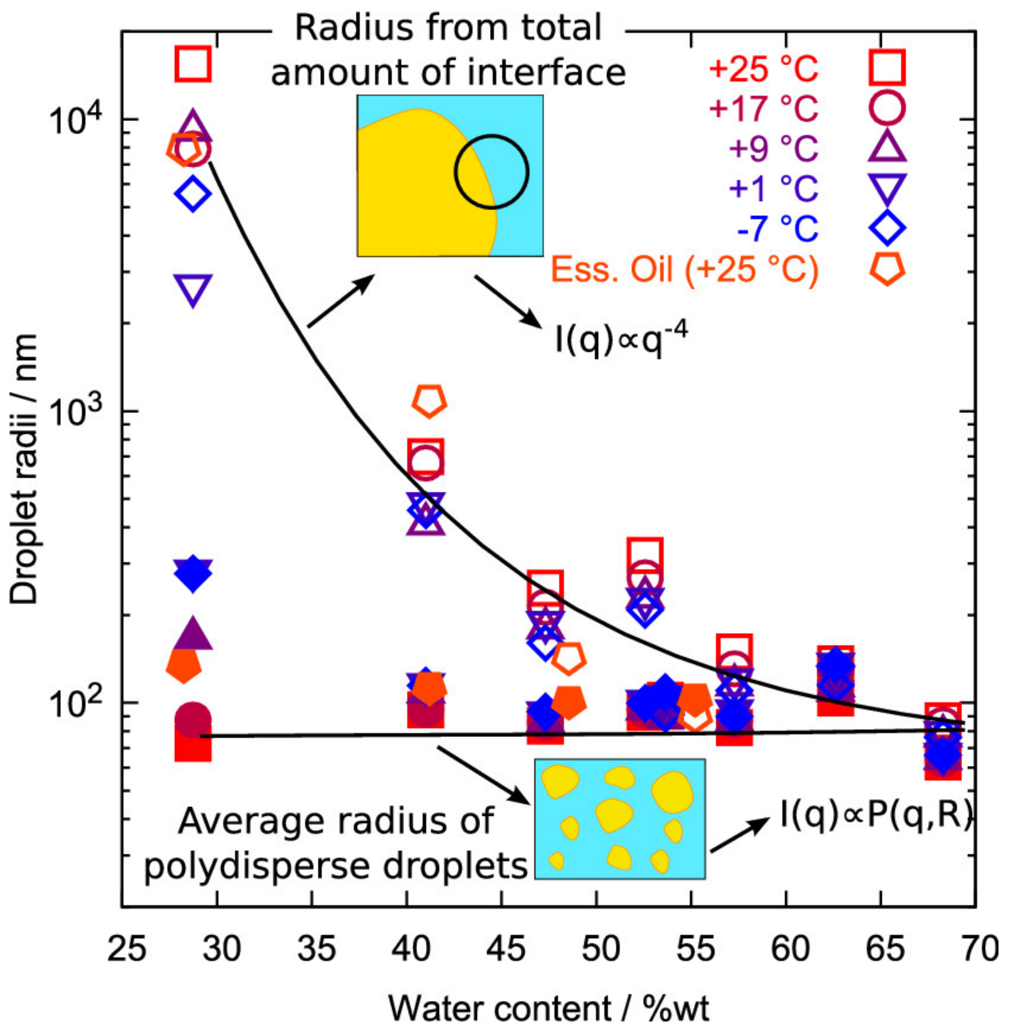
Scattering from Limoncello

- The size of droplets in Limoncello with added water are constant (~100 nm). This is smaller than for other ouzo effect drinks.
- The authors suppose that this is due to the complex mixture of essential oils in Limoncello.
- The small size of droplets is consistent with Limoncello being turbid but having long-term stability.



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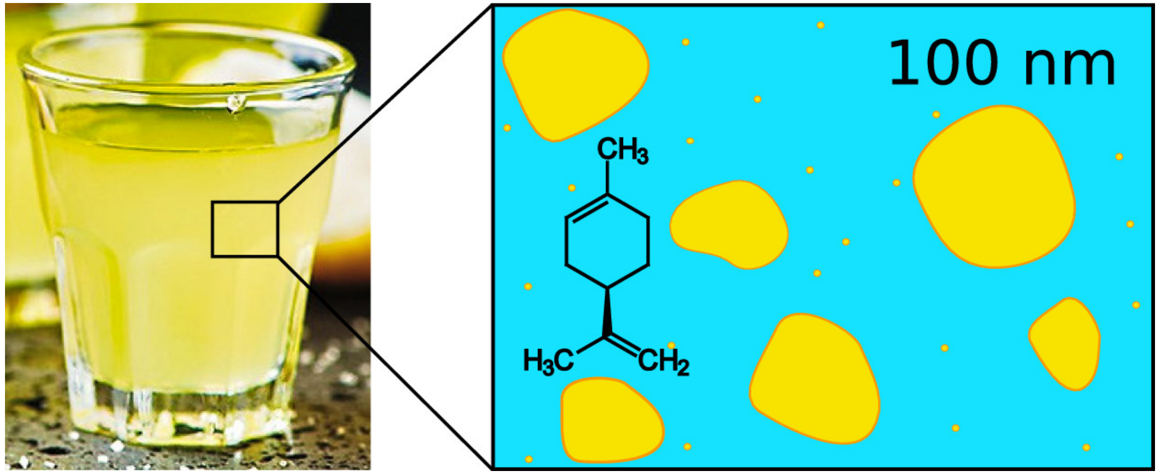
Looking into Limoncello

- There was a strong justification for using neutrons.
- The authors understood how they needed to introduce contrast into the system to get useful information.
- They simplified the system enough that the material can be made, but not so much that the results are no longer meaningful.

To do this well takes thought, effort and planning.



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Objectives

1

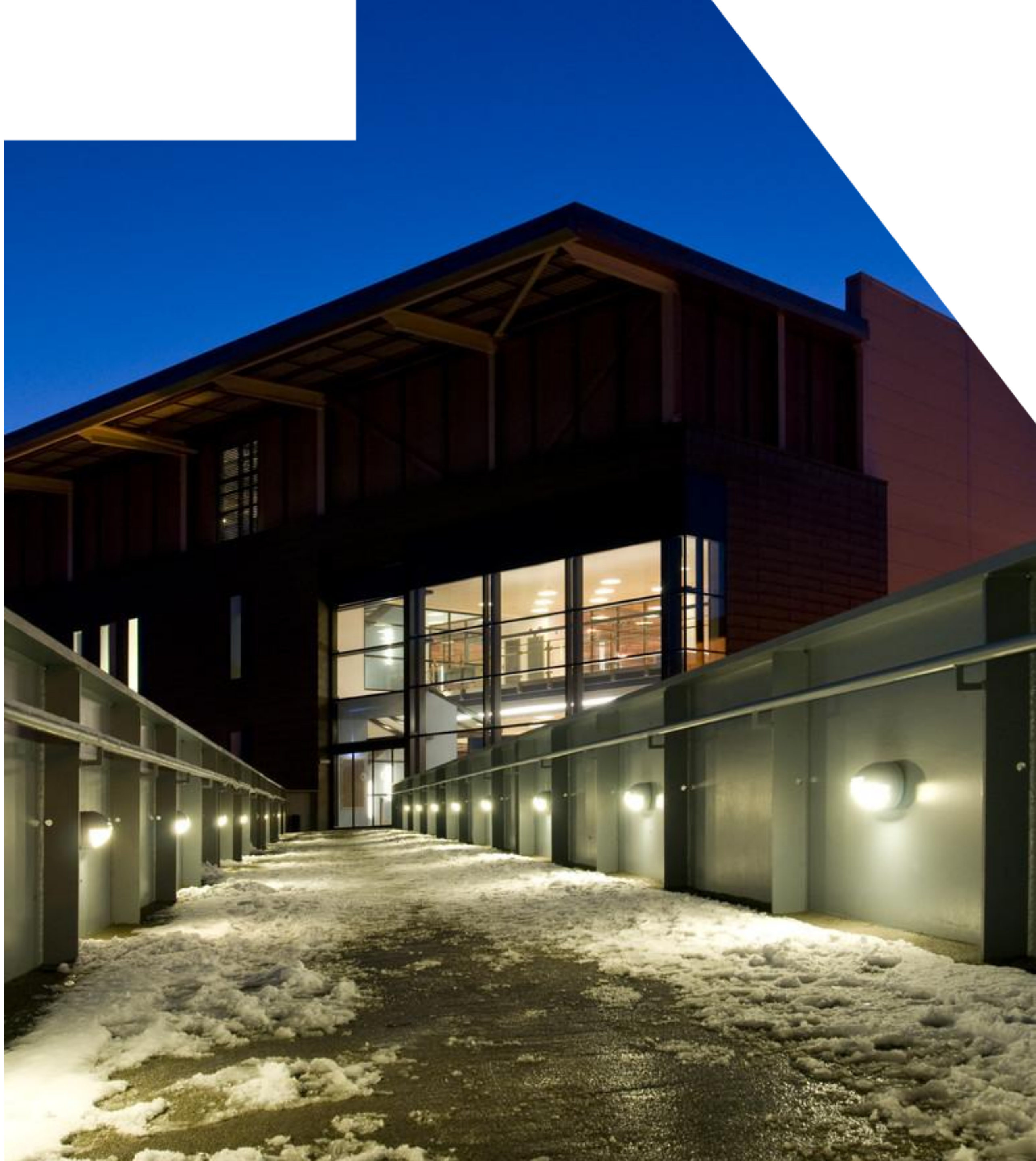
Understand how neutrons and X-rays interact differently with materials and why they are used in investigations

2

Be familiar with large-scale neutron and X-ray facilities and different types of instruments

3

Be aware of past investigations using neutrons and X-rays to study food



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Thank you

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Questions?