PAUL SCHERRER INSTITUT

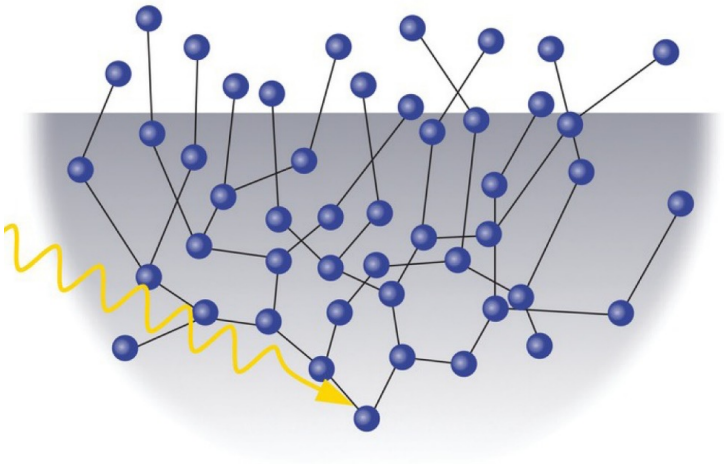
PSI

Materials Science at Large Scale Facilities

Photon Emission Electron Microscopy

Photoemission electron microscopy

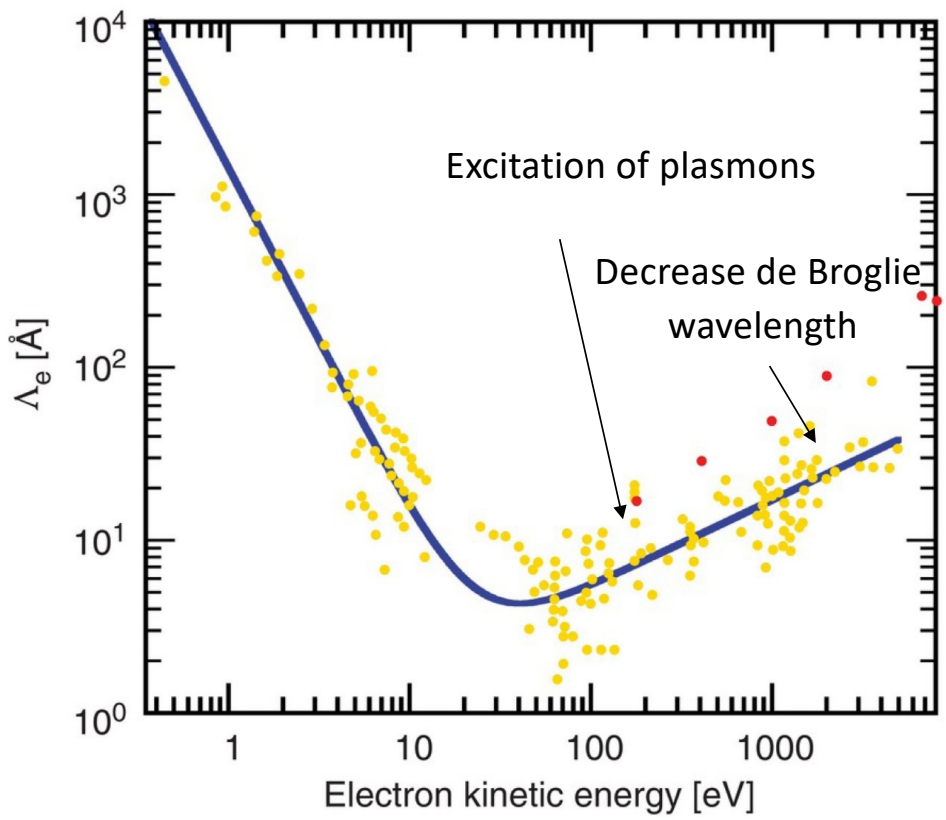
- Photoemission electron microscopy (PEEM) is a technique that images the spatial distribution of electrons emitted from a sample by x-ray absorption
- Although the technique uses photoelectrons for its signal, it is not an electron-spectroscopy technique, as the energies of the electrons are not distinguished.
- The absorption coefficient is indirectly measured via the yield of *secondary* electrons – not the photoelectrons directly emitted after photoabsorption but the electrons released by the system after multiple scattering events in a cascade process originating with the directly produced photoelectron from the absorbing atom. This initial photoelectron may derive from an atom relatively deep in the material, as the x-rays can penetrate to depths of several tens of nanometres.



Philip Willmott, An Introduction to Synchrotron Radiation, 2019

Photoemission electron microscopy

Inelastic mean free path of electrons in matter



Philip Willmott, An Introduction to Synchrotron Radiation, 2019

Region A — High energy (> 200 eV):

IMFP increases because fast electrons interact weakly (small cross-section).

Region B — Mid energy (~20–200 eV):

IMFP is **minimum**

→ strong plasmon excitation

→ lots of inelastic scattering

Region C — Low energy (< 10 eV):

IMFP rises again

→ plasmons cannot be excited anymore

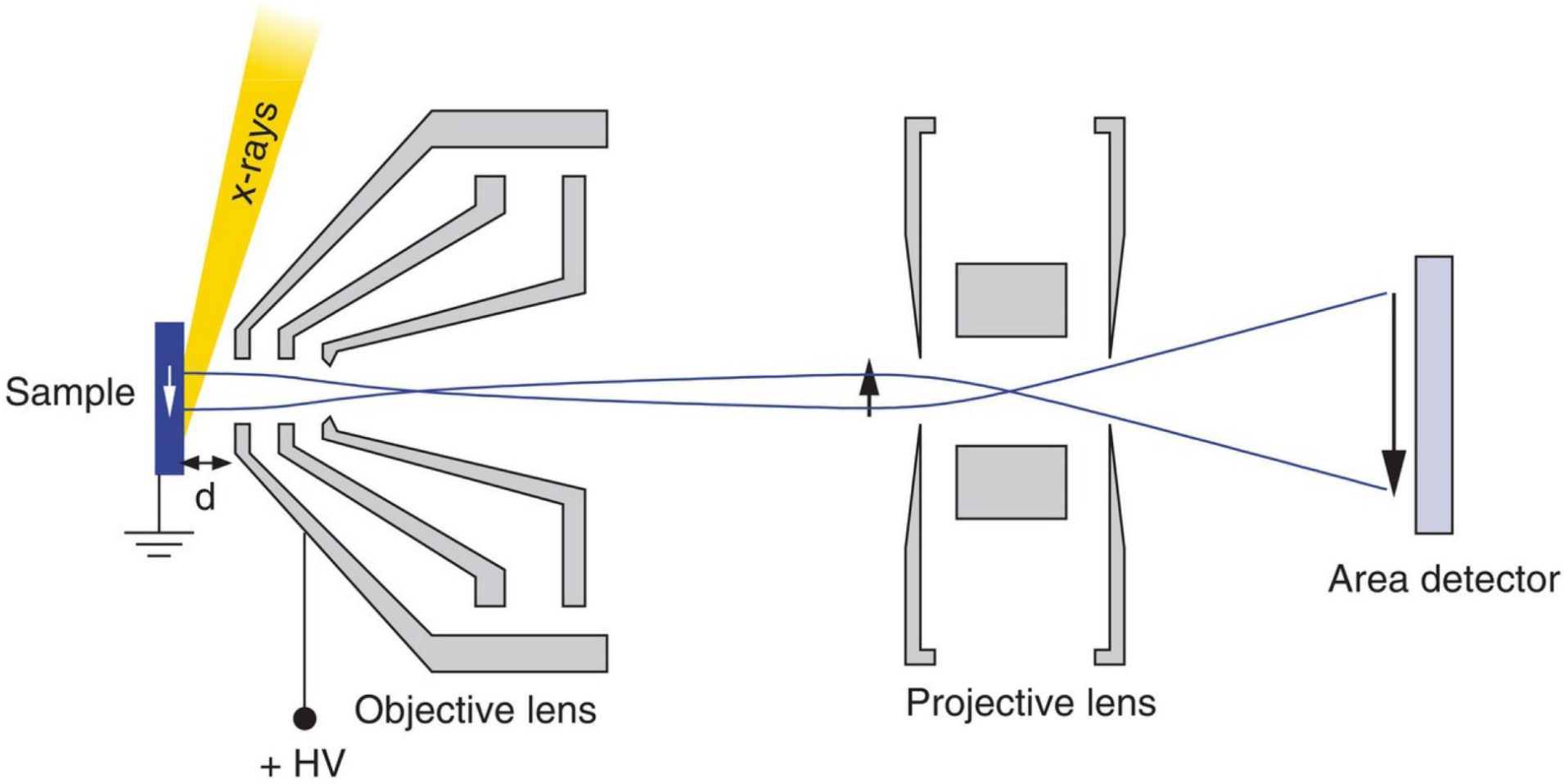
→ not enough electron energy to trigger most inelastic events**

Photoemission electron microscopy

- A PEEM experiment essentially consists of tuning synchrotron radiation illuminating an entire sample in vacuum and imaging the spatial variation of the subsequent secondary-electron yield using an electrostatic and/or electromagnet lens system very similar to that used in electron microscopes
- PEEM is a form of ‘spectromicroscopy’ – full-field images are recorded at different photon energies, in contrast to STXM, a XANES microspectroscopy technique in which a focussed x-ray beam is rastered across the sample.
- The secondary electrons are imaged using a 2D detector. The magnified image of the surface can therefore be observed directly and in real time.

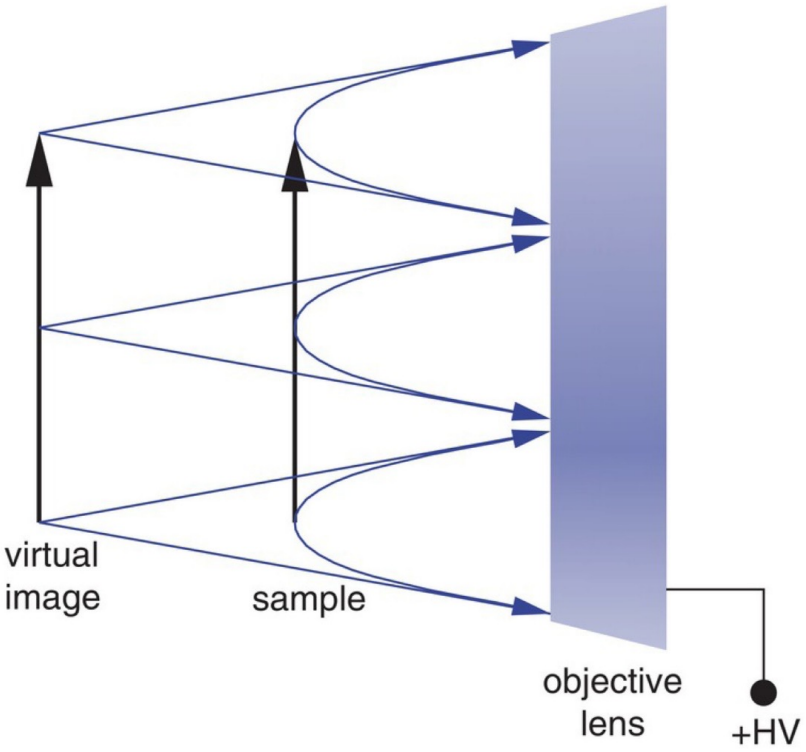
Philip Willmott, An Introduction to Synchrotron Radiation, 2019

Photoemission electron microscopy



Philip Willmott, An Introduction to Synchrotron Radiation, 2019

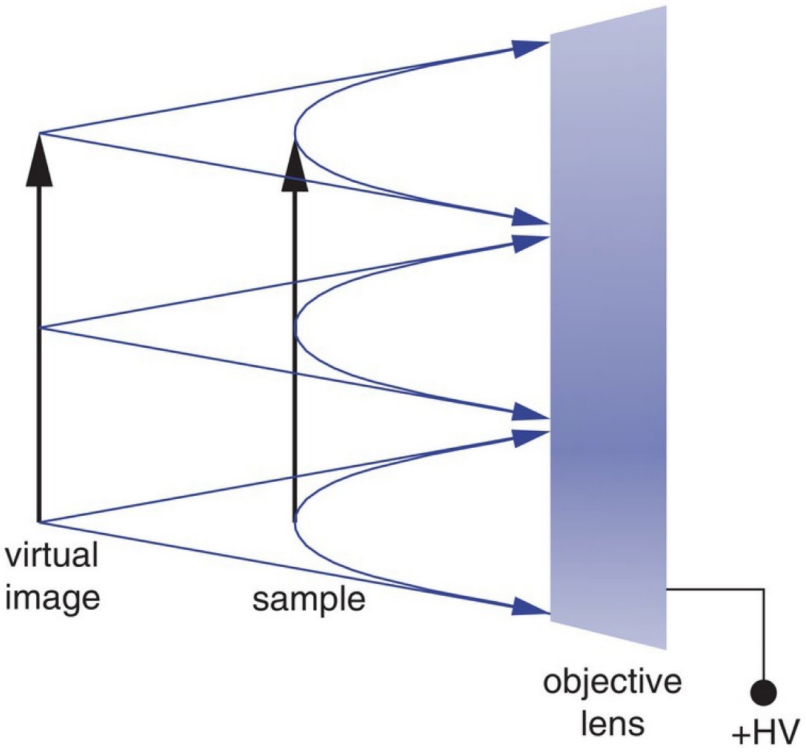
Resolution



- Since electrons are used for imaging in PEEM, the resolution is not limited by the wavelength of the x-ray photon beam.
- A high electrostatic field between the sample and the objective lens accelerates the released electrons to energies of typically eV_L = 10 to 20 keV across a distance d of the order of 2 mm. This accelerating field acts as a lens – the trajectories of the electrons as they leave the surface form a set of parabolas.
- The tangents to the parabolas at the point where the electrons enter the objective lens extrapolate back to form a virtual image with unity lateral magnification.

Philip Willmott, An Introduction to Synchrotron Radiation, 2019

Resolution



- The accelerating field is critical in determining the ultimate resolution. Because the electrons have a range of energies and emission angles, the virtual image will become blurred to a greater or lesser extent.
- The range of electron energies $\Delta\mathcal{E}_e$ can be reduced by introducing an aperture between the sample and objective. This limits the largest parabola width that can enter the electron microscope and therefore acts as a low-pass energy filter, though obviously at the expense of signal intensity.
- To a first approximation, the resolution Δx is given by

$$\Delta x = \frac{d\,\Delta\mathcal{E}_e}{eV_L}.$$

- The best achievable lateral resolution is therefore about 20 nm.

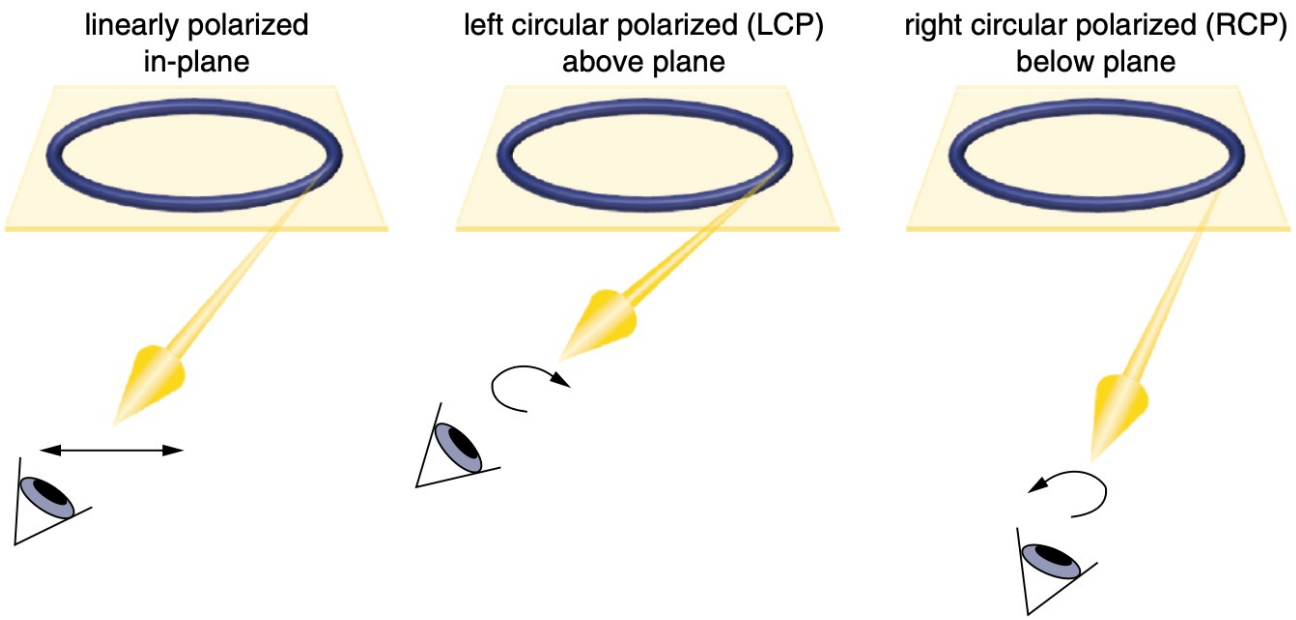
Philip Willmott, An Introduction to Synchrotron Radiation, 2019

PEEM and Magnetic Dichroism

- Dichroism, meaning ‘two-coloured’, is the phenomenon of a material having an absorption spectrum which changes according to the polarization of the electromagnetic radiation used.
- Magnetic dichroism describes the dependence of the absorption of a magnetic material on the polarization *and* the relative orientation of an applied external magnetic field.
- Two methods:
  – x-ray magnetic circular dichroism (XMCD)
  – x-ray magnetic linear dichroism (XMLD)

Philip Willmott, An Introduction to Synchrotron Radiation, 2019

Polarization



The angular momentum of the rotating electric field of the LCP photons is $-\hbar$, and is described by a vector of that magnitude pointing opposite to the direction of propagation. The opposite is true for RCP.

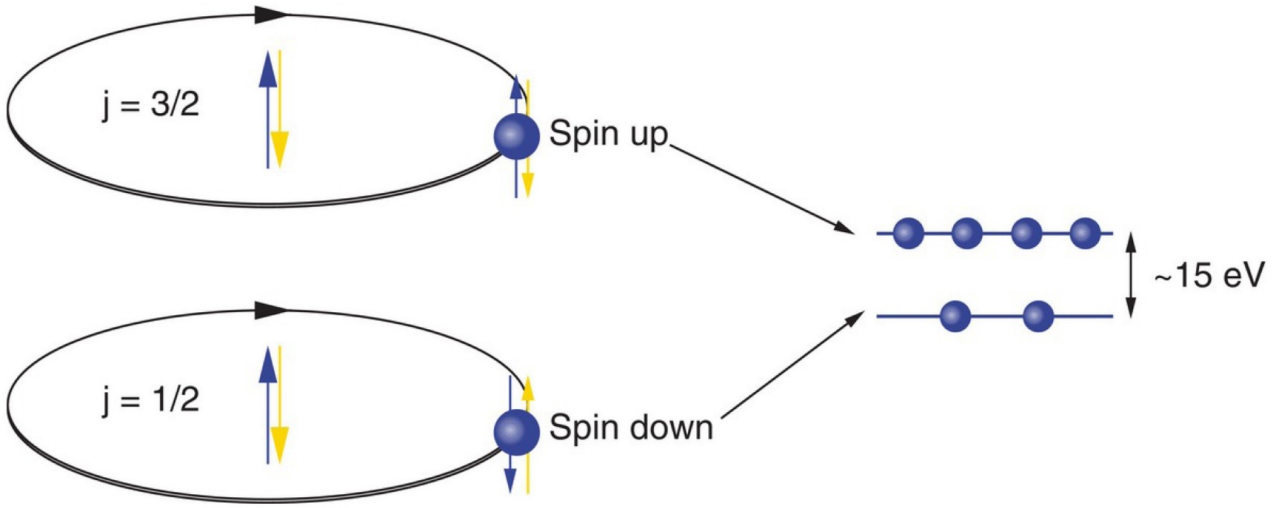
Philip Willmott, An Introduction to Synchrotron Radiation, 2019

First some magnetism

- The intrinsic **magnetic moment** of an electron is produced by its **spin** and can be thought of as being a tiny bar magnet. The orientation of the spin relative to a magnetic field (either +1⁄2, ‘up’, or −1⁄2, ‘down’) determines its only two possible orientations.
- Bound electrons in atoms which have non-spherically symmetric orbitals (i.e. not *s*-type) also have an **orbital angular momentum**, *l*, and therefore generate *a second magnetic field* that can be thought of classically as being produced by the ‘current’ of the electron as it orbits the atom.
- These magnetic moments of the spin and the orbital angular momenta couple with each other just as two magnets are influenced by each others’ fields, in so-called ‘spin-orbit coupling’. Depending on whether the spin is oriented up or down relative to the axis of the orbital magnetic moment, the energy of the electron is higher or lower

Philip Willmott, An Introduction to Synchrotron Radiation, 2019

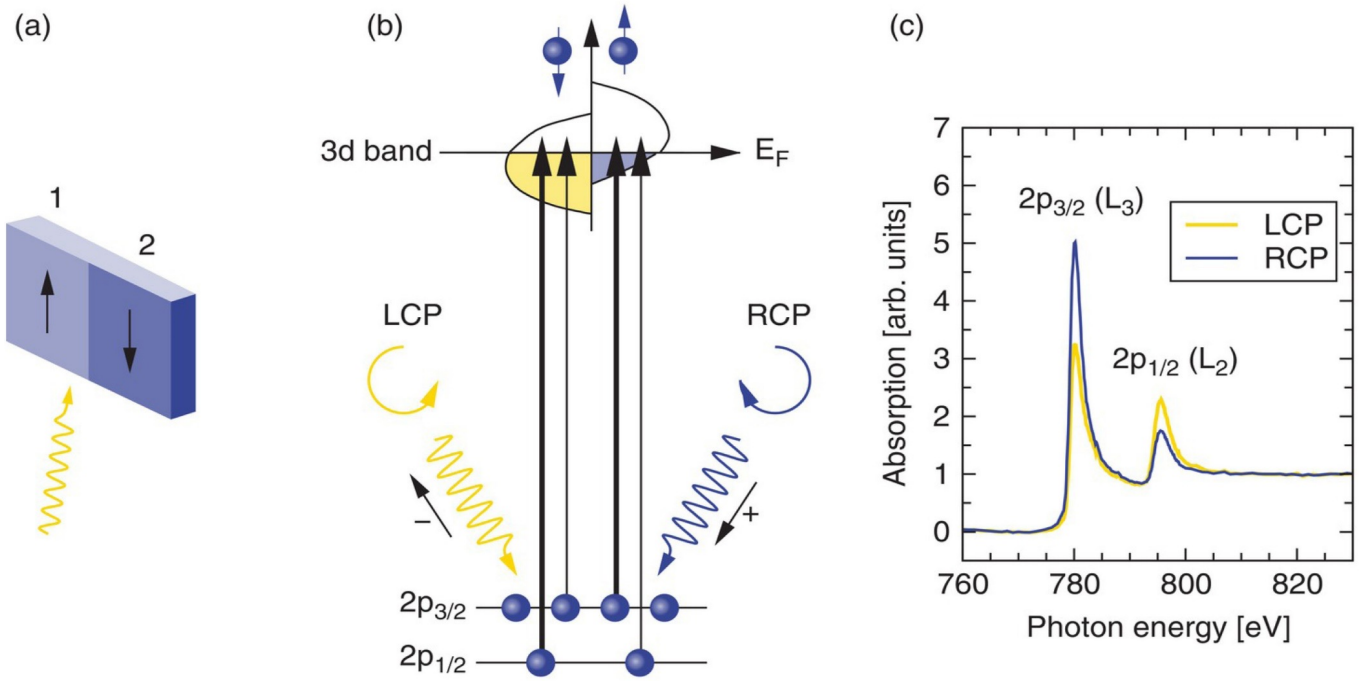
First some magnetism



An example of spin-orbit coupling. The 2*p* orbital has an orbital angular momentum *l* = 1 and an associated magnetic moment which couples to that of the spin of the electron, causing the level to split into two distinct eigenstates *j* = 1 + 1⁄2 = 3⁄2 and *j* = 1 − 1⁄2 = 1⁄2. When these angular momenta are parallel, the magnetic moments are aligned unfavourably compared to when they lie antiparallel to each other. Hence *j* = 3⁄2 has a higher energy than *j* = 1⁄2. The maximum allowed occupation of each state is determined by the number of quantum-mechanically allowed projections *mj* of *j* relative to the magnetic axis and is equal to 2*j* + 1. The magnetic moments, shown in yellow, are antiparallel to their associated angular momenta, shown in blue.

Philip Willmott, An Introduction to Synchrotron Radiation, 2019

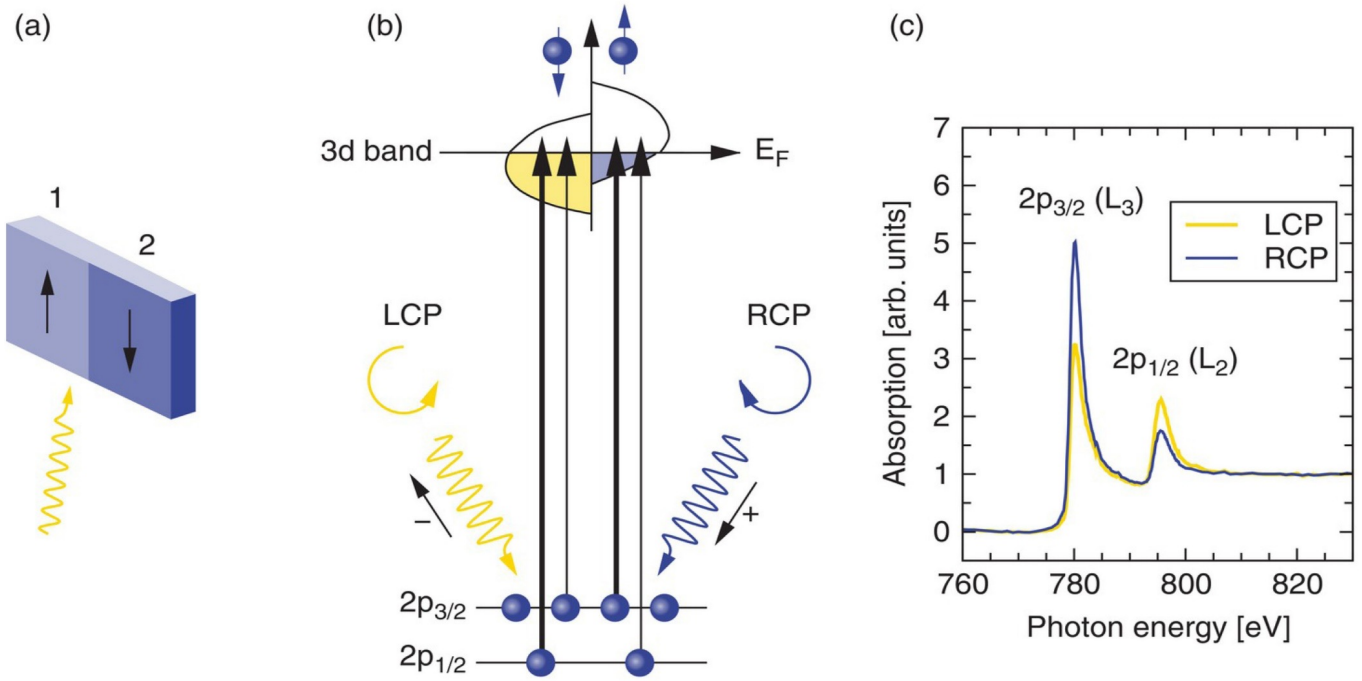
X-ray magnetic circular dichroism - XMCD



- A ferromagnetic material such as cobalt with 3d valence electrons forms domains with different magnetic directions. In region 1, there are more occupied 3d states with their spin down than with spin up, hence the magnetization of this domain is upwards.
- In contrast, the number and density of unoccupied spin-up states above the Fermi level in the 3d band is greater than spin-down states for region 1 and the transition probability for absorption of a photon will be greater.

Philip Willmott, An Introduction to Synchrotron Radiation, 2019

X-ray magnetic circular dichroism - XMCD



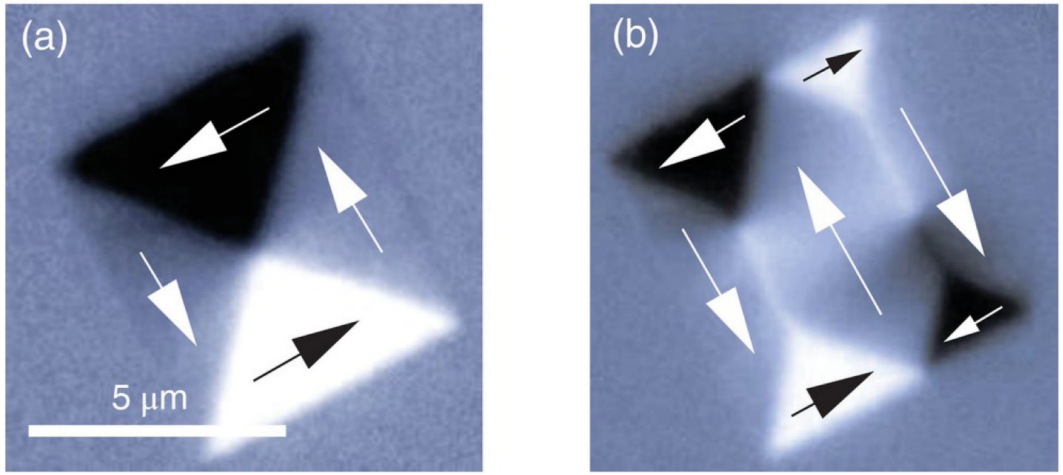
- In the case of region 1, an incident RCP photon has its orbital angular momentum pointing forwards (in the direction of propagation) and will preferentially excite the $2p_{3/2}$ (L3) levels over the $2p_{1/2}$ (L2) levels. The opposite case arises for LCP photons.
- For region 2, the whole story reverses

Absorption of RCP photons mainly excites spin-up electrons, while LCP light mainly excites spin-down electrons, resulting in a dichroic absorption spectrum

Philip Willmott, An Introduction to Synchrotron Radiation, 2019

X-ray magnetic circular dichroism - XMCD

- By recording PEEM images of ferromagnetic domains with RCP and LCP x-rays and dividing the intensities of one image, pixel-for-pixel, by those of the other, the domain structure is revealed with maximum contrast.

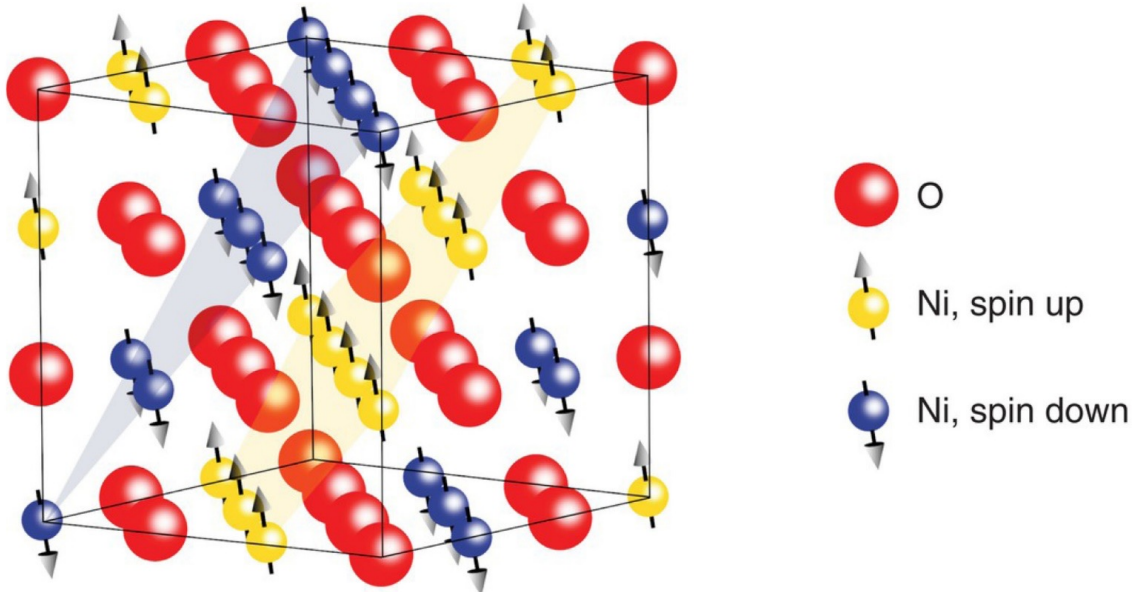


The orientation of microscopic domains in Ni-Fe thin films minimize the stray field energy. Such domains can break up into higher-energy metastable configurations

Philip Willmott, An Introduction to Synchrotron Radiation, 2019

X-ray magnetic linear dichroism - XMLD

- A technique used to investigate the domain structure of *anti*ferromagnetic (AFM) materials.
- XMLD signal arises from the fact that, due to spin-orbit coupling, the spatial distribution of the electron density is marginally distorted, providing the necessary dichroism depending on whether the linear polarization vector is parallel to or perpendicular to the magnetic axis.



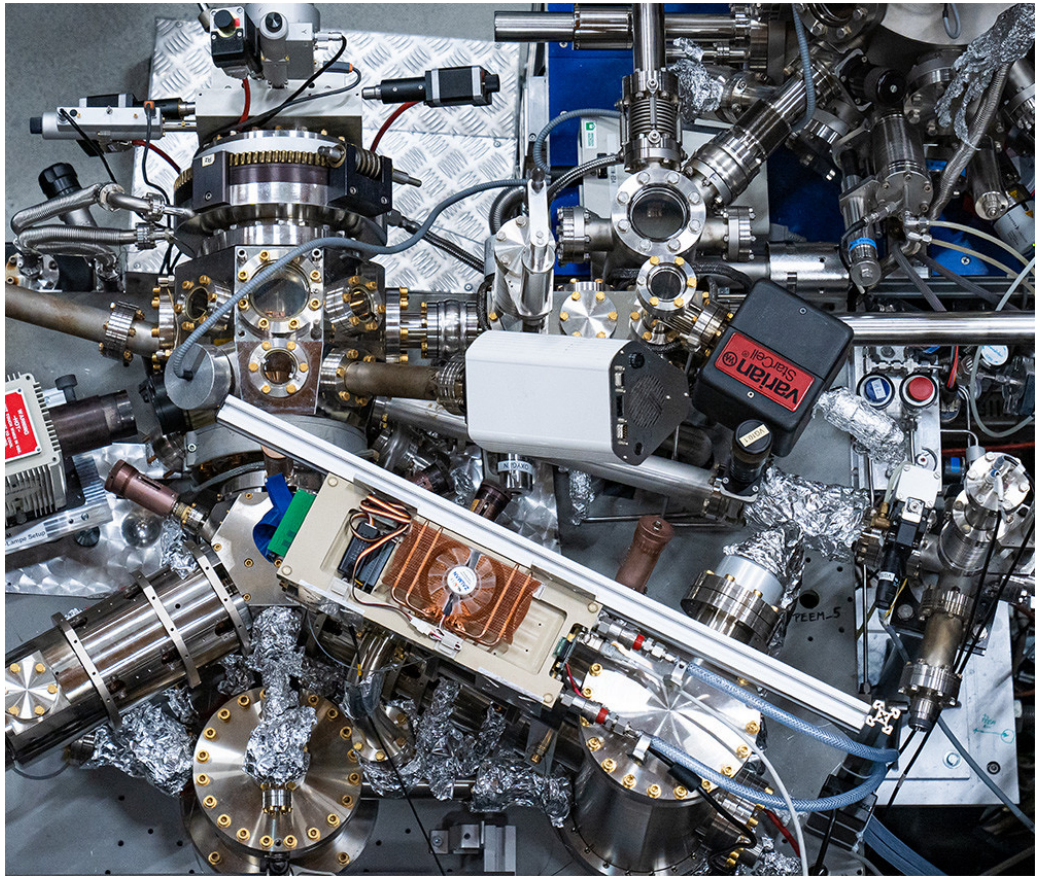
NiO is an antiferromagnetic cubic crystal with a face-centred cubic rocksalt structure. Below the Néel temperature, the spins of the nickel ions in the (111) crystallographic planes (shown here in blue and yellow) alternate between being all spin-up and all spin-down. There is therefore no net magnetic moment, but there does exist a magnetic axis, given by the spin orientations.

Philip Willmott, An Introduction to Synchrotron Radiation, 2019

PEEM @ SLS

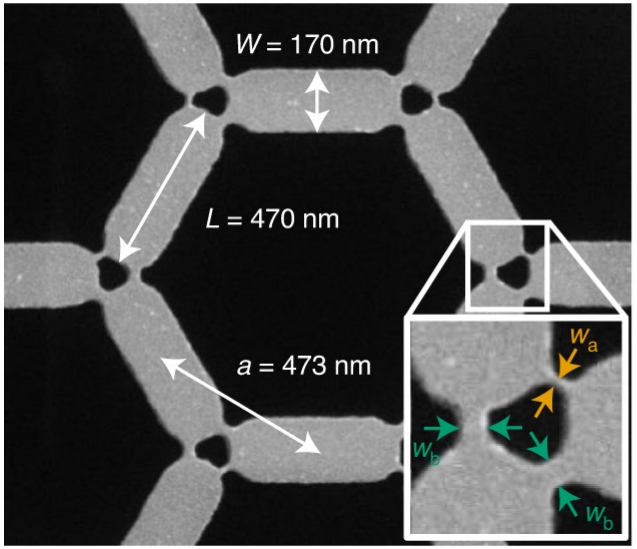
- Surfaces / Interfaces: Microscopy (SIM)

Spatial resolution	down to ~50 nm
Electron energy resolution	0.2 eV
Sample temperature	120 K < T < 1'800 K
Field of view	5 - 150 µm diameter



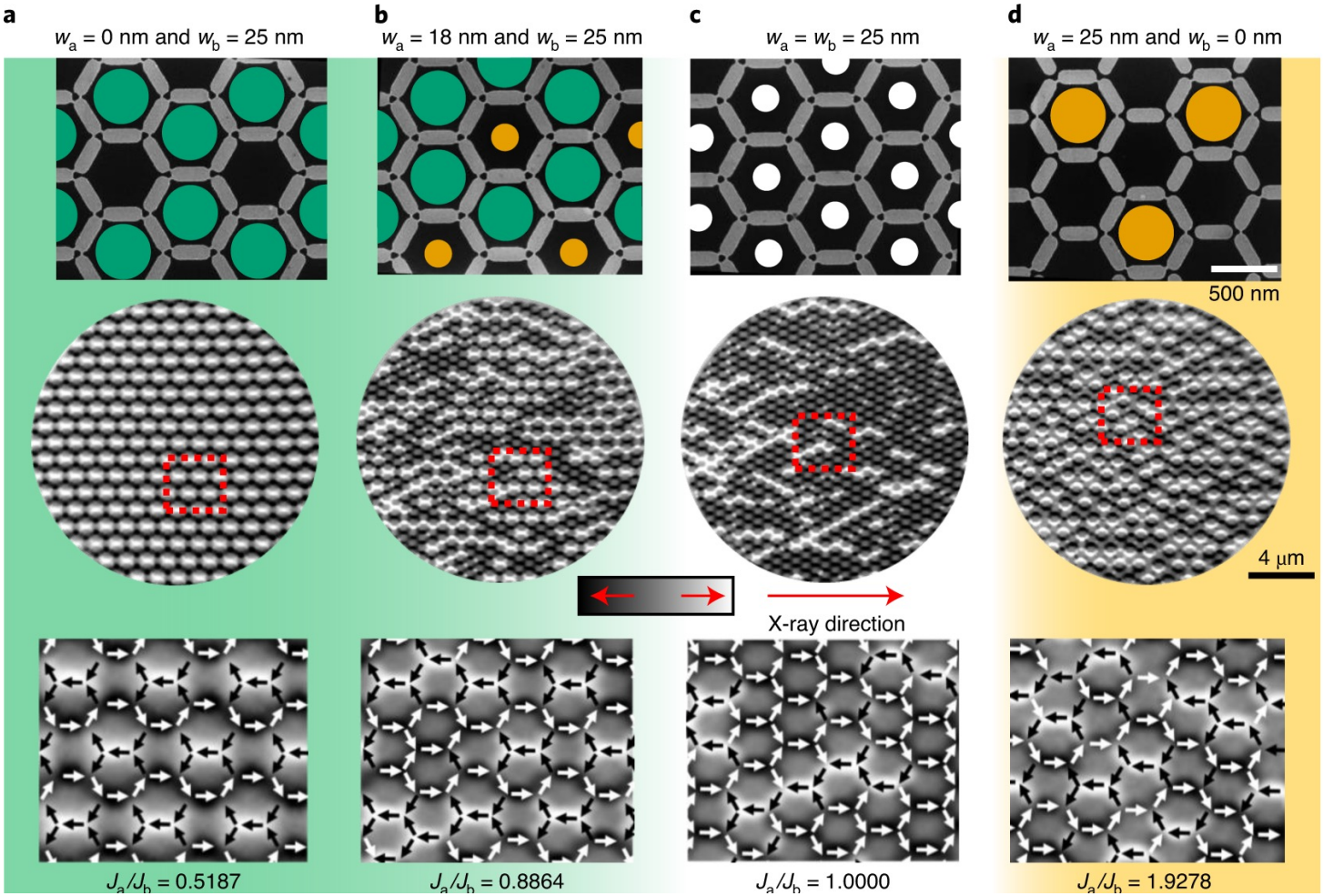
Application: kagome spin ice

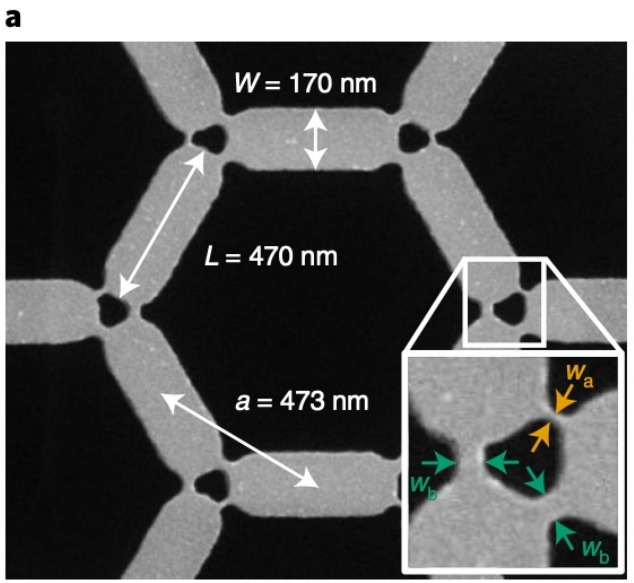
- Artificial spin ice, which essentially consists of nanomagnets that are so small that their orientation can only change as a result of temperature
- The nanomagnets are arranged in hexagonal structures – a pattern that is known from the Japanese art of basket weaving under the name kagome.
- A nickel-iron compound called permalloy, which was coated as a thin film on a silicon substrate. Lithography was used to repeatedly form a small, hexagonal pattern of nanomagnets, with each nanomagnet being approximately half a micrometer.
- Goal: observe phase formation as a function of temperature



K. Hofhuis, Nature Physics **18**, (2022) 699–705

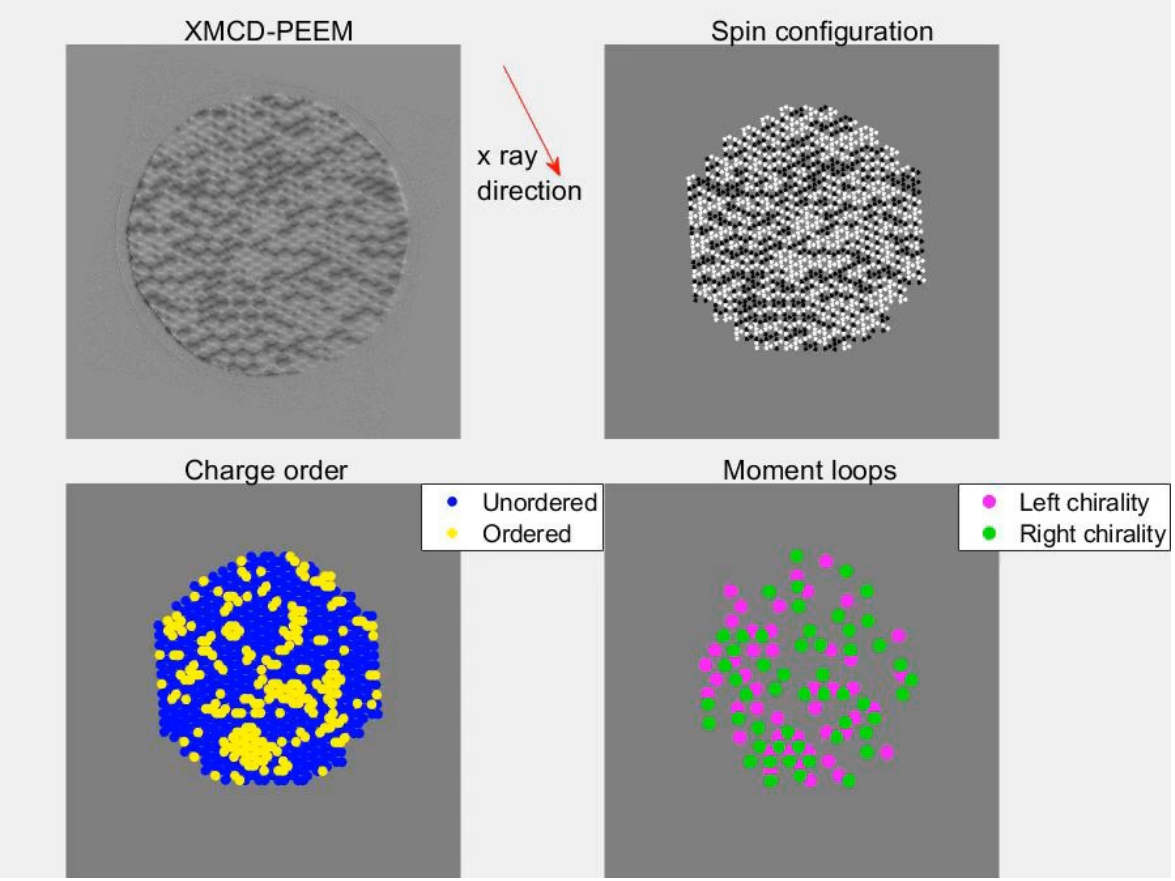
Application: kagome spin ice





K. Hofhuis, Nature Physics **18**, (2022) 699–705

Application: kagome spin ice



K. Hofhuis, Nature Physics **18**, (2022) 699–705