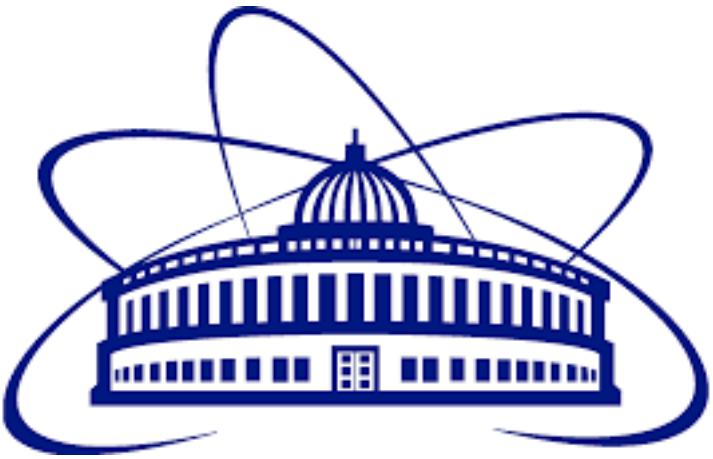

Introduction to neutron scattering experiments at large scale facilities.

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Abstract:

Neutron scattering is a unique tool for research studies of the fundamental properties observed in condensed and soft matter. Small-angle scattering (SAS) is the collective name given to the techniques of small-angle neutron (SANS), X-ray (SAXS) and light (SALS, or just LS) scattering. In each of these techniques, radiation is elastically scattered by a sample and the resulting scattering pattern is analysed to provide information about the size, shape and orientation of some components of the sample. SasView is an open-source project currently hosted on Github. The software is written in Python with C and C++ modules providing some of the heavier computations. This report will highlight some of the basic notions of neutron scattering experiments and will make use of the SasView software package for the visualization of experimental data.

Introduction:

Neutrons:

A neutron is a subatomic particle with a mass equal to 1.67×10^{-27}kg, with no charge and with a magnetic moment of -1.93μN where μN is a nuclear magneton. It is a fermion with intrinsic angular momentum equal to 1/2ħ where ħ is reduced Planck constant and it was discovered in 1932 by James Chadwick. Together with protons, it constitutes nuclei of an atom. Atoms of a chemical element that differ only in neutron number are called isotopes. Protons and neutrons are attracted to each other via the strong force. Since their discovery in 1932 neutrons play an important role in many fields of modern science. The discovery of the neutron immediately gave scientists a new tool for probing the properties of atomic nuclei. In particular, the discovery of neutrons and their properties has been important in developing nuclear reactors and nuclear weapons. A nuclear reactor is a key device of nuclear power plants, nuclear research facilities, or nuclear-propelled ships. The main purpose of the nuclear reactor is to initiate and control a sustained nuclear chain reaction. The nuclear chain reaction is initiated, sustained, and controlled just via the free neutrons. **The term chain** means that one single nuclear reaction (neutron-induced fission) causes an average of **one or more** subsequent nuclear reactions, thus leading to the possibility of a self-propagating series of these reactions. The “one or more” is the key parameter of

reactor physics. To raise or lower the power, the amount of reactions, respectively the amount of the free neutrons in the nuclear core, must be changed (using the control rods). **Neutron diffraction** experiments use an **elastic neutron scattering** to determine the atomic (or magnetic) structure. The neutron diffraction is based on the fact that thermal or cold neutrons have wavelengths similar to **atomic spacings**. An examined sample (crystalline solids, gasses, liquids, or amorphous materials) must be placed in a neutron beam of thermal (0.025 eV) or cold (neutrons in thermal equilibrium with very cold surroundings such as liquid deuterium) neutrons to obtain a diffraction pattern that provides information about the structure of the examined material. The neutron diffraction experiments are similar to X-ray diffraction experiments, but neutrons interact with matter differently. Photons (X-rays) interact primarily with the electrons surrounding (atomic electron cloud) a nucleus, but **neutrons interact only with nuclei.**

Neutron Sources:

There are two means of producing neutrons in sufficient quantities for worthwhile experiments. The most obvious of these is to use a nuclear reactor. Here neutrons are released by the fission of uranium-235. Each fission event releases 2 - 3 neutrons, though one of these is needed to sustain the chain reaction. The most powerful of the reactor (or what are also termed "steady-state" or "continuous") neutron sources in the world today is the 55 MW HFR (High-Flux Reactor) at the Institute Max von Laue - Paul Langevin (usually shortened to just the "ILL") in Grenoble, France.27 The ILL is jointly operated by France, Germany, the United Kingdom, and 12 other countries. The facility commenced operation in 1972.

The other approach to neutron production is that used in spallation neutron sources. These use particle accelerators and synchrotrons to generate intense, high-energy, proton beams which are, in turn, directed at a target composed of heavy nuclei. Provided that the protons have sufficient kinetic energy they are able to overcome the intrinsic long-range electrostatic and short-range nuclear forces they encounter and effectively blast the target nuclei apart. The word spallation is a quarrying term for "chipping away". The most successful spallation neutron source in the world is the ISIS Facility near Oxford in the United Kingdom.28-30 It is based around a 200 A, 800 MeV (ie, 160 kW), proton synchrotron operating at 50 Hz, and a tantalum target which releases approximately 12 neutrons for every incident proton. ISIS is operated by the United Kingdom but also receives funding from a variety of international partners. The facility commenced operation in late 1984. The most powerful spallation neutron source in the world is the 850 kW SNS at the Oak Ridge National Laboratory, USA.

Today there are some 37 neutron sources in 21 countries31; of these, 23 are in continental Europe (including Russia and Scandinavia), 9 are in North America (including Canada; IPNS closed in 2008), 2 are in Japan with 1 in each of Australia and India. Five of the sources are spallation sources, the remainder are generally rather aging reactors, though

some, such as the ILL, have undergone recent refurbishment to extend their useful lifetimes.32 The total number of SANS instruments at these sources is 36; of which 21 are to be found at the European facilities.33 Despite this apparent glut of facilities and instruments, the demand for SANS beam time typically outstrips the beam time actually available by a factor of 2 or 3.

Small Angle Neutron Scattering:

Small-angle neutron scattering (SANS) is one of the most important techniques for microstructure determination, being utilized in a wide range of scientific disciplines, such as materials science, physics, chemistry, and biology. The reason for its great significance is that conventional SANS is probably the only method capable of probing structural inhomogeneities in the bulk of materials on a mesoscopic real-space length scale from roughly 1 to 300 nm. Moreover, the exploitation of the spin degree of freedom of the neutron provides SANS with a unique sensitivity to study *magnetism and magnetic materials* at the nanoscale. As such, magnetic SANS ideally complements more real-space and surface-sensitive magnetic imaging techniques, e.g., Lorentz transmission electron microscopy, electron holography, magnetic force microscopy, Kerr microscopy, or spin-polarized scanning tunneling microscopy. This review summarizes the recent applications of the SANS method to study magnetism and magnetic materials. This includes a wide range of materials classes from nano magnetic systems such as soft magnetic Fe-based nano composites, hard magnetic Nd-Fe-B-based permanent magnets, magnetic steels, ferrofluids, nanoparticles, and magnetic oxides to more fundamental open issues in contemporary condensed matter physics such as skyrmion crystals, non collinear magnetic structures in non centrosymmetric compounds, magnetic or electronic phase separation, and vortex lattices in type-II superconductors. Small angle scattering (SAS) is the collective name given to the techniques of small angle neutron (SANS), X-ray (SAXS) and light (SALS, or just LS) scattering. In each of these techniques radiation is elastically scattered by a sample and the resulting scattering pattern is analysed to provide information about the size, shape and orientation of some component of the sample.

The type of sample that can be studied by SAS, the sample environment that can be applied, the actual length scales that can be probed and the information that can ultimately be obtained, all depend on the nature of the radiation employed. For example, LS cannot be used to study optically opaque samples and SAXS cannot (easily) be employed to study thick samples or samples requiring complex containers, whilst SANS (and SAXS) probe different length scales to LS.

Neutron reflectometry experimental facilities:

Neutron reflectometry needs reflection of neutrons with a momentum transfer in the range from 0.001-1 Å-1. There are two ways to obtain the necessary values of the momentum transfer: the first is to carry out measurements by very cold neutrons at large angles of incidence, the second is to use neutrons with wavelengths in the few angstroms range at small angles of incidence of the neutron beam (~ 0.1° -2°) with low divergence.

Neutron reflectometers can be divided into three classes:

1.Time-of-flight

2.Monochromatic

3.Combined. Facility works in time-of-flight and monochromatic mode at the same time.

Monochromatic reflectometers operate on continuous neutron sources. They provide a higher resolution on the momentum transfer than time-of-flight reflectometers. Monochromators usually made from pyrographite. It is necessary to scan the angle of incidence and scatter (θ-2θ-geometry) to measure R(Q). The typical value of the operating wavelength is 4-5 Å, i.e. close to the maximum of the reactor spectrum. Time-of-flight reflectometers work both on pulsed neutron sources and on continuous ones. In pulsed neutron sources the neutron beam is modulated by choppers. Usually scanning is carried out by wavelength, i.e. by the time of neutron flight with a stationary sample and detector.

All reflectometers have a similar structure. Their main parts:

1. Neutron guide.

Mirror neutron guides have been installed between the source and the installations for increasing the flux density of thermal and especially cold neutrons. Curved neutron guides or straight ones with a curved section are also widely used. The simplest mirror neutron guide is a pipe with polished walls made from highly scattering material. The guides were evacuated to prevent the neutrons from scattering on the air molecules, and the low divergent neutrons hitting the neutron guide walls at low grazing angles were reflected and transported more efficiently. Most materials have a refractive index that is slightly below 1 for cold and thermal neutrons. To reflect neutrons at still higher angles than that corresponding to m=1, one could use the ^{58}Ni isotope. This has a higher neutron scattering length density compared to Ni, equivalent to m=1.18. The material with the highest neutron scattering length density in the periodic table is carbon in the form of a diamond. Thereafter come Ni and Be, which have nearly the same scattering length density. Both C and Behave with a very low absorption cross section compared to Ni, and when a neutron is absorbed, both emit gamma radiation at lower photon energies than Ni.

2. Collimating devices (slits).

Slits provide the necessary divergence of the beam and the dimensions corresponding to the sample. Usually, collimating devices are made with a changeable slit size.

3. Deflecting mirror.

It is used in time-of-flight reflectometers with a vertical scattering plane. Allows you to set the incidence angle of the neutron beam leaving the sample plane horizontal. The position of the mirror is set using a goniometer that moves the mirror in a vertical plane. Super mirrors or polarizing mirrors (in reflectometers with a polarized beam) are used as a deflecting mirror.

4. Polarizer and spin-flipper.

Magnetic mirrors made from ferromagnets (for example, Co) or magnetic super mirrors are used as polarizers. Magnetic super mirrors are becoming increasingly popular because they work in a wider range of momentum transfer. The spin-flipper is used to reverse the polarization of neutrons. Neutron radio frequency (RF) spin- flippers with mutually opposing currents are also used in neutron reflectometers. These spin flippers provide a high degree of polarization in a wide range of neutron wavelengths.

5. Sample part.

It includes a displacement device that moves the sample in three mutually perpendicular directions and a goniometer that rotates or swings the sample in the scattering plane within a few degrees and with a precision of at least 1'.

6. Polarisation analyser.

Allows you to separate neutrons reflected with a spin flip from neutrons reflected without a spin flip. by design. Polarised 3He has been used as polarizers and analyzers. These devices provide a wide aperture, high polarisation efficiency in a wide range of wavelengths and can work in beams with high divergence. However, this type of polarizers requires periodic laser pumping. Laser pumping systems are very bulky and expensive.

The mirror analyser allows you to register neutrons reflected with a spin flip and without a flip at the same time. The detector registers neutrons reflected from the analyzer mirror and those that have passed through it. This scheme allows you to decrease the measurement time if you need to analyse the polarisation after reflection by the sample.

7. Detector.

The detector is necessary for measuring the reflected neutrons and flux that determines the neutron reflection coefficient. It is necessary to measure the angular distribution of the scattered neutrons in the scattering plane to study non-mirror reflections caused by the irregular surface and axial geometry relative to the axis of the incident beam for studies of (GISANS). Two-coordinate detector is required for this case. The coordinate resolution of the detectors is 1-3 mm for each coordinate, the typical efficiency of registration is about

30%. Usually, the detectors are placed in a vacuum tube, inside which it can move, or the detector rotates with the tube, thus changing the viewing angle. The use of a vacuum tube helps to avoid neutron losses due to scattering and absorption in air.

SasView:

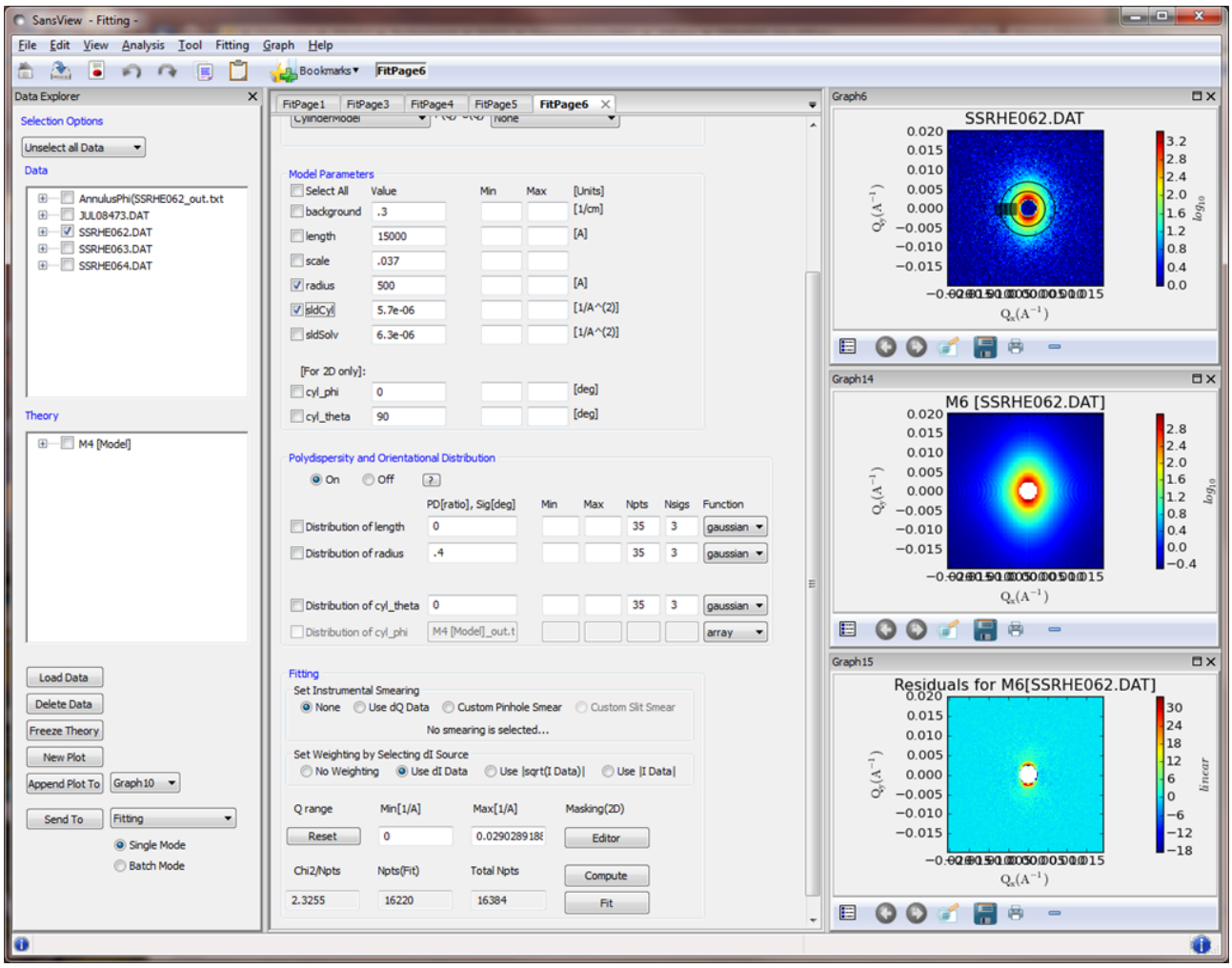
SasView is an open-source project currently hosted on GitHub. The software is written in Python with C and C++ modules providing some of the heavier computations. Persons interested in becoming developers are welcome and even encouraged. Have a look at how you can get involved or contact the project leadership team at management@sasview.org. Pull requests can also be made for one-off contributions. Comments, suggestions, and other contributions are also welcome and can be made by emailing the development team at developers@sasview.org.

SasView was originally developed by the University of Tennessee as part of the Distributed Data Analysis of Neutron Scattering Experiments (DANSE) project funded by the US National Science Foundation (NSF) but is currently being developed as an Open Source project hosted on GitHub and managed by a consortium of scattering facilities. Participating facilities include (in alphabetical order): the Australian National Science & Technology Centre for Neutron Scattering, the Diamond Light Source, the European Spallation Source, the Federal Institute for Materials Research and Testing, the Institut Laue Langevin, the ISIS Pulsed Neutron & Muon Source, the National Institute of Standards & Technology Center for Neutron Research, the Oak Ridge National Laboratory Neutron Sciences Directorate, and the Technical University Delft Reactor Institute.

SAS analysis is a fairly broad term, potentially covering a huge amount of ground and often meaning different things to different people. As part of this project, several boundaries were identified and defined. The first boundary is the distinction between SAS reduction and SAS analysis. Reduction is defined as the process that removes all instrument-specific “artefacts” and produces a reduced data set containing only the “science” of the material being investigated. If done perfectly reduced data from any instrument should lie on top of one another with minor caveats regarding differences in the contrast term arising out of the use of different types of radiation. The rationale is that every instrument is different and every source is different. Thus reduction will always be a process relatively unique to the instrument on which the data was taken and only the facilities’ instrument scientists are truly qualified to know what all those details are. By contrast, once in reduced form, the job of the user scientist can begin in earnest as that is the data that contains the information he/she is seeking. Further, at this point, the “analysis” problem becomes a completely common and generic problem so that all facilities and the community at large can easily collaborate having now the exact same

needs. There are grey areas where the distinction is not as clear. However, we believe in most cases the problem can be reduced to this separation. SasView is designed to operate on **reduced** data and thus should be instrument agnostic.

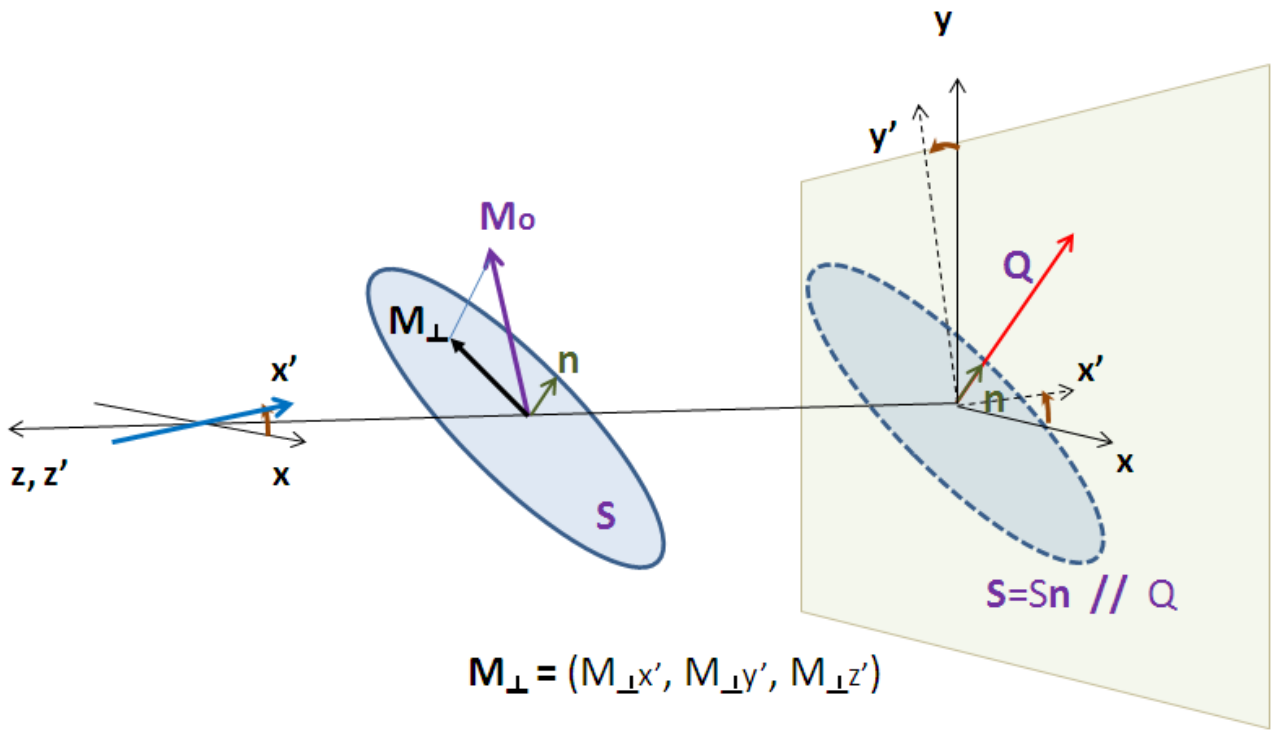
The second demarcation is between real space and inverse space analysis. By inverse space analysis we mean the process of thinking about data in inverse space as collected, while by real space analysis we mean the process of thinking about the data in real 3D space. The first case represents the traditional approach to SAS analysis used by colloid and materials scientists while the second has been pioneered mostly by the biological scattering community. We believe that these are two very different (if complementary) ways of thinking about the problem and it would be very difficult to create a single user interface ideal for both. Thus SasView is designed as an inverse space modelling tool. We hope to build partnerships to interact with real space analysis packages such as the SASSIE project, providing if appropriate support in making them relevant beyond biological systems. Two methods cross this boundary: Distance distribution, P(R), and correlation function analyses which attempt a direct inversion of the data to real space with no a priori assumptions, and shape reconstruction or “ab initio” methods which seek to refine an arbitrary 3D real space model (again with no a priori knowledge) till its Fourier inversion fits the scattering data. The first method is incorporated into SasView. At this time the second is thought to be more appropriate for the real space analysis packages. The philosophy is to try to balance the desire to do all one’s analysis in one package with the need to keep the interface somewhat intuitive and easy to learn and use. The hope is to achieve this balance by packaging all the functionality likely to be wanted by most users of this package most of the time and to design and build interfaces with complementary packages to allow data to be moved transparently across applications when needed.



2D fitting screenshot

Magnetic Scattering:

For magnetic scattering, only the magnetization component, M⊥, perpendicular to the scattering vector Q contributes to the magnetic scattering length.



The magnetic scattering length density is then

$$\beta_M = \frac{\gamma r_0}{2\mu_B}\sigma \cdot \mathbf{M}_\perp = D_M \sigma \cdot \mathbf{M}_\perp$$

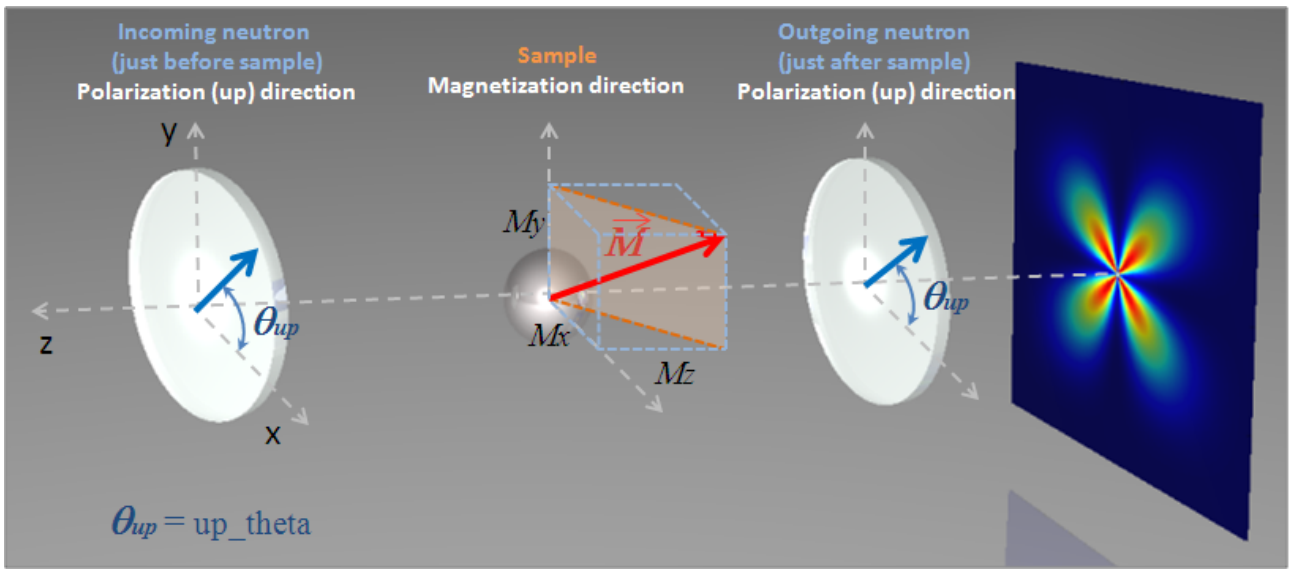
where the gyromagnetic ratio is γ = −1.913γ=−1.913, B μB is the Bohr magneton,r0

is the classical radius of the electron, and σ is the Pauli spin.

For a polarized neutron, the magnetic scattering is depending on the spin states.

Let us consider that the incident neutrons are polarised both parallel (+) and anti-parallel (-) to the x' axis. The possible states after scattering from the sample are then

- Non-spin flips: (+ +) and (- -)
- Spin flips: (+ -) and (- +)



Now let us assume that the angles of the Q vector and the spin-axis (x') to the x-axis are φ and θ up respectively (see above). Then, depending upon the polarization (spin) state of neutrons, the scattering length densities, including the nuclear scattering length density (βN) are given as

- for non-spin-flips
$$\beta_{\pm\pm} = \beta_N \mp D_M M_{\perp x'}$$

- for spin-flips
$$\beta_{\pm\mp} = -D_M \left(M_{\perp y'} \pm i M_{\perp z'}\right)$$

where

$$M_{\perp x'} = M_{0q_x} cos\theta_{up} + M_{0q_y} sin\theta_{up}$$
$$M_{\perp y'} = M_{0q_y} cos\theta_{up} - M_{0q_x} sin\theta_{up}\, M_{\perp z'} = M_{0z}$$
$$M_{0q_x} = (M_{0x} cos\phi - M_{0y} sin\phi) cos\phi$$
$$M_{0q_y} = (M_{0y} sin\phi - M_{0x} cos\phi) sin\phi$$

Here the

M0 x, M0y and M0z are the x, y and z components of the magnetisation vector in the laboratory xyz frame.

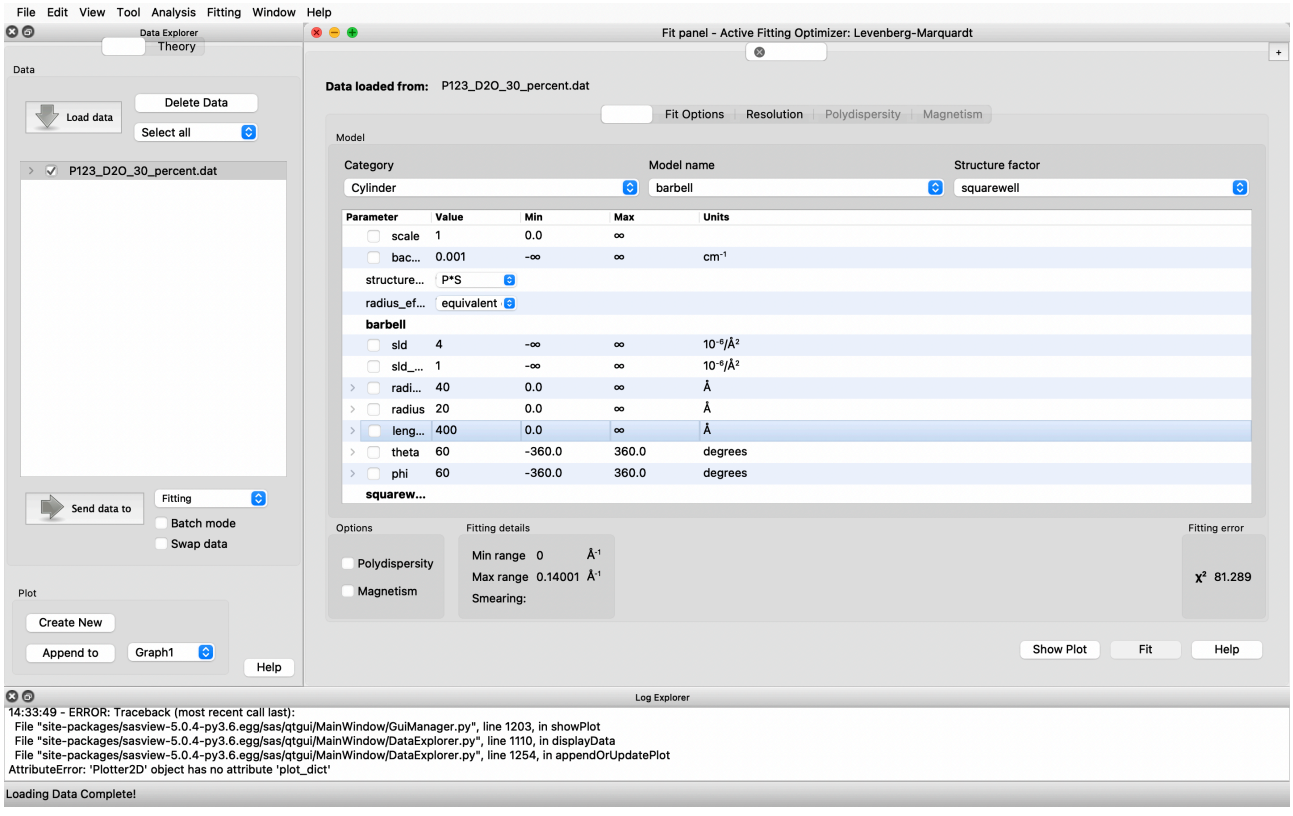
Magnetic SANS and Data Analysis:

We begin this section with a description of a typical SANS setup the basic expressions for the various unpolarized and spin-polarized elastic SANS cross-sections $d\Sigma/d\Omega$ will be displayed. We focus on the two most relevant scattering geometries which have the applied magnetic field H0 either perpendicular or parallel to the wave vector k^0 of the incoming neutron beam. In the first Born approximation, the magnetic contribution to $d\Sigma/d\Omega$ is fully determined by the three Cartesian Fourier. By means of a mechanical velocity selector or copper-based time-of-flight methods, the incoming wave-length band (typically $\lambda \sim 3 - 30$ ˚A) is selected from a cold neutron beam [energy range: $\sim 0.1 - 10$ meV $\sim 1 - 120$ K (Schober, 2014)], provided by spallation or a reactor source. The mean wavelength and wavelength resolution can be tuned [$\Delta\lambda/\lambda \sim 1 - 30$ % (FWHM)], depending on the rotational speed and tilting angle of the selector or the duty cycle and frame overlap of the chopper system. In the evacuated pre-sample flight path a set of apertures Two-dimensional position-sensitive detector arrays, moving along rails in an evacuated post-sample flight path (sample-to-detector distance: $\sim 1 - 40$ m), count the scattered neutrons during acquisition times ranging between a few minutes and a few hours. The recorded neutron counts (in each pixel element) are corrected for detector dead time, dark current and efficiency, sample transmission and background scattering and are normalized to incident-beam flux. A solid-angle correction is applied to the data which corrects for the planar geometry of the detector. The size of an individual pixel element of the detector is typically 10 mm×10 mm, so that the related resolution effects become negligible. The scattering cross section of the sample is obtained by comparing the corrected signal to a reference sample (e.g., water, polystyrene, porous silica, vanadium single crystal) of the known cross section. The data reduction procedure provides the macroscopic differential scattering cross section $d\Sigma/d\Omega$ of the sample collimates the beam. A particular strength of the SANS technique is that experiments can be conducted under rather flexible sample environments in absolute units (typically cm^{-1}) and as a function of the magnitude and orientation of the momentum-transfer or scattering vector q (see Fig. 1). In order to conveniently present the neutron data, one often carries out a so-called azimuthal averaging procedure, whereby the data at a constant magnitude of q are integrated within a certain angular range (e.g., over $0 \leq \theta \leq 2\pi$); this yields $d\Sigma/d\Omega$ as a function of $|q| = q$. The uncertainty in the cross sections determined by this procedure is estimated to be $5 - 10$ %.

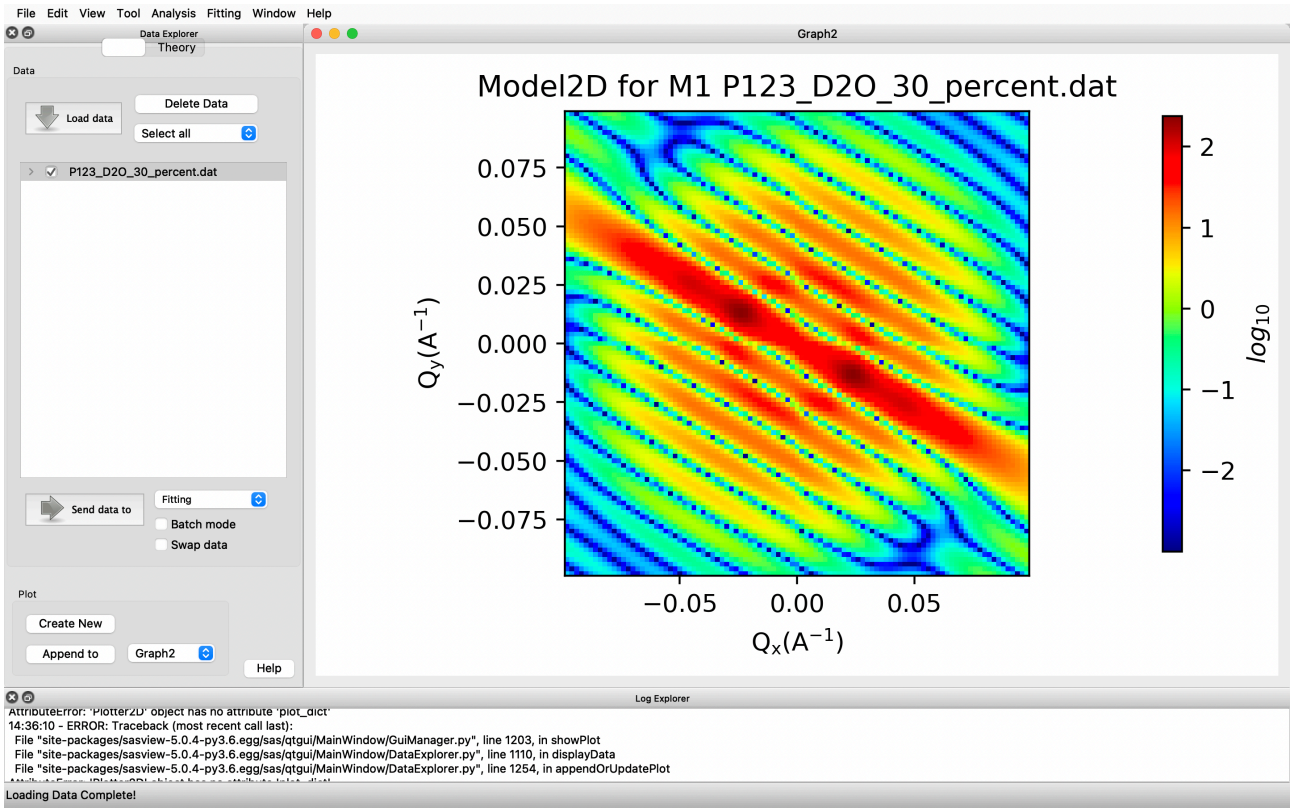
The relative significance of coherent and incoherent neutron scattering depends on the nuclear composition of the sample, the size of the particles or structures present in the sample, and on the range of neutron wave vector transfer (Q) accessed in an experiment. In a small-angle neutron scattering (SANS) experiment Q values are small, and, given a sufficient scattering contrast, coherent scattering from large objects dominates the scattering pattern even when these objects have many hydrogen nuclei (which have a high incoherent scattering cross section). With increasing Q, coherent scattering drops fast (following for instance Guinier's Law) and often becomes much smaller than the

incoherent component. Relative to SANS, in a conventional quasielastic neutron scattering (QENS) experiment the Q values are high (>0.2Å–1Å), the molecules are often small, and the hydrogen content is high enough to reduce the coherent scattering contribution to a few percent and less. This is why an analysis of QENS experiments often accounts for incoherent scattering only (see, for instance,
In a SANS diffraction experiment, incoherent scattering is just a flat background, whereas coherent scattering is a source of structural information. In QENS, incoherent scattering informs us about the single-molecule motion, and the motions of individual functional groups within the molecule, while coherent scattering gives us information about the motion of molecules (and their parts) relative to each other. Hence, coherent QENS is more difficult to analyze than incoherent. First, the dependence of the line shape of the coherent QENS spectra on the structural properties of the sample is more intricate; consequently, much of the structural information (e.g., the solute's crystal structure, radial distribution functions in solution) is required as model input. Second, it is in general much more difficult to model the collective motion of a system of particles, than the motion of a single particle.

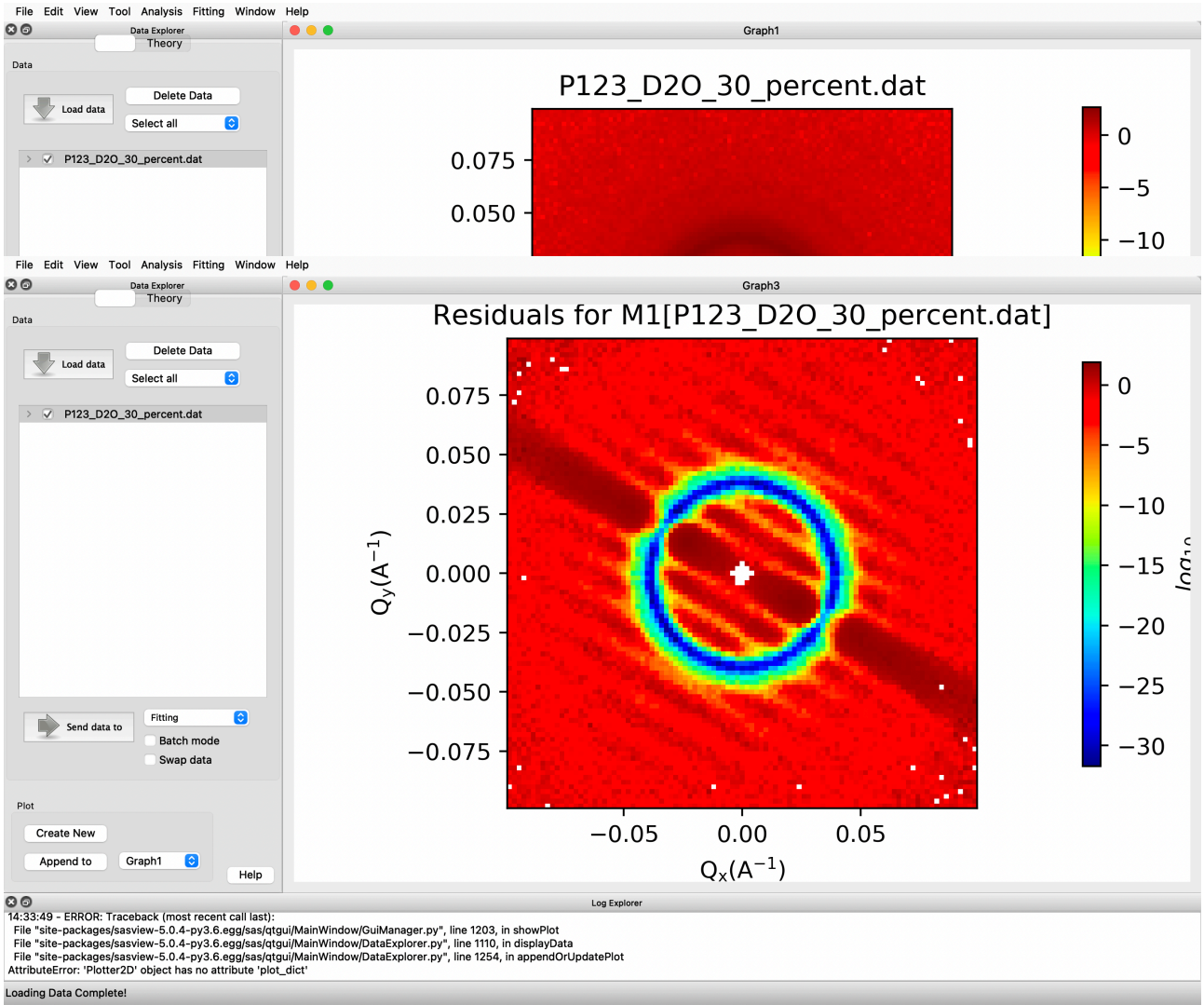
2D Data Analysis for D2O 30% Solution.



P123_D2O_30% data



2D model for P123_D2O 30% solution data.



Residuals for M1 30% D2O data.

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