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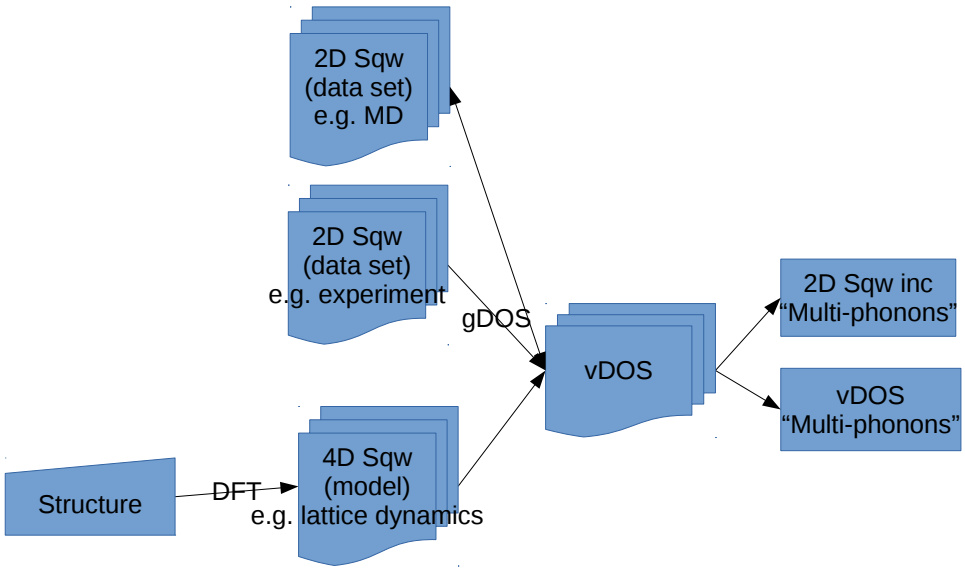
Introduction

It is well-known in the field of neutronic simulations [see papers by F. Cantargi, R. Granada, JI Márquez Damián et al below] that the cross section descriptions e.g. in the meV range distributed via NDEF [NDEF/NJOY] could be improved.

Hence, the underlying rationale for the algorithm described below is to make use of available materials theories like MD or DFT as well as experimental neutron scattering data from e.g. ToF spetrometer studies, thereby improving realism of cross-section descriptions for neutronic simulations.

Elements of material structure and dynamics



Knowing a material to be used in nuclear facilities allows to measure and/or compute a number of physical quantities. The chemical formula (and space group) allows to compute the structure factor, which represents the spatial arrangement of atoms in space. In addition, that same information can be used to get an estimate of the density of states using a molecular/atomistic modelling. This information quantifies the typical vibrational energies of movements in the material, which are responsible for the moderation of neutrons.

The neutron scattering technique permits to measure the structure factor, using neutron diffraction, and the phonon spectrum, using inelastic spectrometers. However, as any measurement, data treatment must be carried out to extract the theoretical information, removing e.g. instrumental effects, including resolution, and additional contributions to the measured signal.

Existing methodologies date from 30 years ago, and thanks to progresses in computational physics, it is now feasible to extract the material physical quantities with improved accuracy by coupling the data treatment with molecular/atomistic and instrument modelling. Then, it is expected to better estimate important data such as the neutron scattering cross section, especially in the low energy region ($\omega < 100$ meV) which determines the spectral characteristics of the final moderation process events.

The proposed algorithm

The first step is to perform an analysis of reduced inelastic scattering data, removing the multiple scattering and the so-called "phonon expansion" terms from the measurements. This step can then provide, from an experimental signal containing "spurions", a realistic estimate of the "true" vibrational density of states (vDOS), some estimate of the structure factor S(q), as well as *S(q,ω)* and *I(q,t)* models.

The basis of this approach are Sjölander (derived into MuPhoCor by Reichardt for the density of states integral), Sköld/Vineyard/Egelstaff approximation, and S(q,w) sum-rules. The multiple scattering would be estimated from a Monte-Carlo McStas model.

The only input is the reduced, experimental *S(q,ω)* or *S(φ,ω)*, and chemical formula (to get masses, cross sections and derive partial DOS).

<u>So called "Phonon expansion":</u>

In the incoherent approximation, the dynamic structure factor can be written as [Sjölander]:

$$S(Q,\omega) = \frac{\sigma_{\text{inc}}}{4\pi}\frac{N}{2\pi\hbar}\exp\left(-\frac{\hbar Q^2}{2m}f(0)\right)\sum_{p=0}^{\infty}\frac{1}{p!}\int_{-\infty}^{\infty}\mathrm{d}t\mathrm{e}^{-\mathrm{i}\omega t}\left(\frac{\hbar Q^2}{2m}f(t)\right)^p$$

with

$$f(t) = \int_{-\infty}^{\infty}\mathrm{d}\omega\frac{g(\omega)}{\omega}\cdot\left(n(\omega)+1\right)\cdot\exp(\mathrm{i}\omega t)$$

Clearly, this expression is Gaussian in shape, and does not include any structural information except the Debye-Waller factor. It only takes as input the vibrational density of states g(□). Even though it is labelled as "phonon expansion" in the literature, the notion of phonon in a purely incoherent model scatterer is surprising. However, some vibrational information is used as input, which is suited for e.g. molecular spectroscopy.

In order to take into account the structure in the *S(q,ω)* model, and recover some aspects of the coherent scattering, we may make use of the Sköld approximation to derive the coherent contribution from the incoherent one:

$$S_{coh}(Q,\omega) = S(Q)S_{inc}\left(\frac{Q}{\sqrt{S(Q)}},\omega\right) - S_{inc}(Q,\omega)$$

which retains the second frequency moment sum-rule. The input is the structure factor S(q) and an incoherent scattering law $S_{inc}(q,\square)$ estimate, as obtained above.

This way, we may obtain a dynamic structure factor estimate for both the coherent and incoherent contributions. The methodology may be used for e.g. isotropic density materials such as liquids, powders and amorphous systems.

We aim to implement a fully automatic procedure as follows, which takes minimal input information:

1. Provide an experimental reduced *S(q,ω)* or *S(φ,ω)* data set, obtained from e.g. Mantid or LAMP.
2. Compute an estimate of the generalised density of states (gDOS), following Bredov/Oskotskii.
3. Split the total gDOS into partial gDOS per atom, using chemical formula (masses and neutron cross sections).
4. **Refine** the gDOS in order to get the "true" vDOS [in the style of MuPhoCor]. This quantity could also be obtained from a MD or DFT simulation, but then usually requires a substantial computational time.
5. Compute the incoherent *S(q,ω)* [following above equations, Sjölander] and multiple orders.
6. Remove the incoherent multi-phonon terms from the experimental data.
7. Estimate the structure factor S(q) from both the energy integration and the *S(q,ω)* 2nd moment. This quantity could also be obtained from a diffraction experiment, or the chemical formula (using CrysFML), or a MD/DFT simulation.
8. Compute an estimate of the coherent *S(q,ω)* from the Sköld approximation, using the above S(q) and the incoherent *S(q,ω)*.
9. Cross-check the experimental data with the estimated *S(q,ω)* model.
10. Extend the experimental data with the coherent+incoherent model, and use it as input into a McStas model to compute the multiple-scattering contribution.
11. Remove the multiple-scattering contribution.
12. **Loop** iteratively to item 2. until convergence.
13. This way we have subtracted the multi-phonon and multiple scattering contributions, derived a model *S(q,ω)* and refined the density of states. In addition, the *I(q,t)* can be estimated for e.g. NSE experiments.
14. Qualify the result by deriving the frequency moments and write an automatic report

This way, 'clean' dynamical and structural information are obtained, which is, a step forward compared to previous approaches (Carpenter/Bellissent/Reichardt). The indicated algorithm is a completely new implementation (not a MuPhoCor port) and will be made available to non-expert users, and integrated into a simple GUI via the iFit package [http://ifit.mccode.org] .

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