Simulating single-crystal inelastic scattering in full phase-space

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China Spallation Neutron Source

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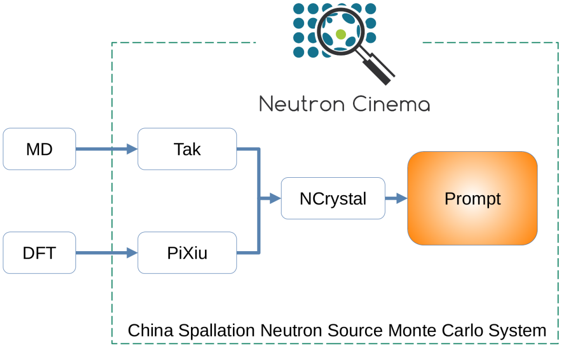
International Collaboration on Advanced Neutron Sources (ICANS XXIV)

1. Cinema
2. Examples
3. PiXiu
4. Sampling

Cinema: China Spallation Neutron Source Monte Carlo System



Neutron Cinema

MD

DFT

Tak

PiXiu

NCrystal

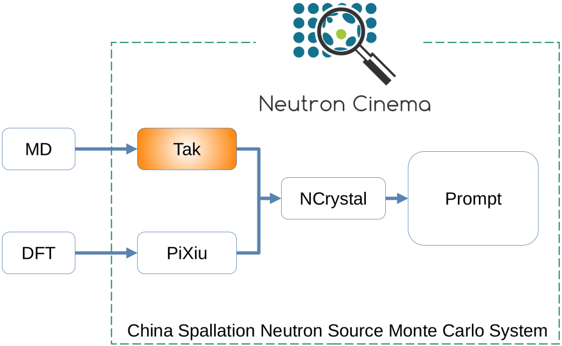
Prompt

China Spallation Neutron Source Monte Carlo System

Prompt, the dual mode Monte Carlo engine:

- Paper @`https://arxiv.org/abs/2304.06226`.
- Source @`https://gitlab.com/cinema-developers/prompt`.
- Installing as `pip install neutron-cinema`.

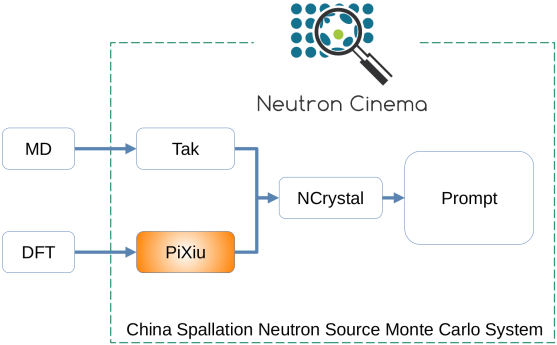
Cinema: China Spallation Neutron Source Monte Carlo System



Tak, Trajectory Analysis Toolkit, bridging the gap between MD and $S(Q,\omega)$:

- Incoherent inelastic calculator published in 2022[R. Du, X.-X. Cai, J. Comput. Phys, 442, 111382, 2022].

Cinema: China Spallation Neutron Source Monte Carlo System



PiXiu, bridging the gap between DFT and $S(\vec{Q}, \omega)$:

- DFT post analysis code for single phonon scattering.
- A reference paper is on preparation.
- Detailed introduction in the talk.

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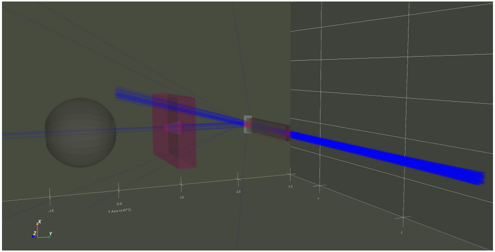
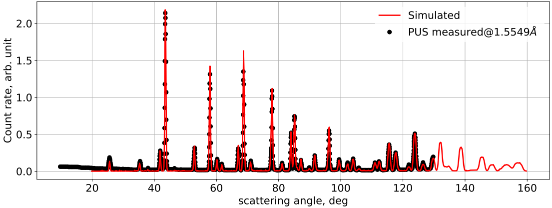
Diffractometer (more examples can be found in the reference paper)

The PUS diffractometer in Norway.

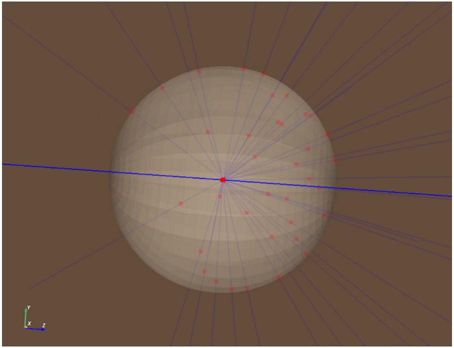




Total scattering

A total scattering setup

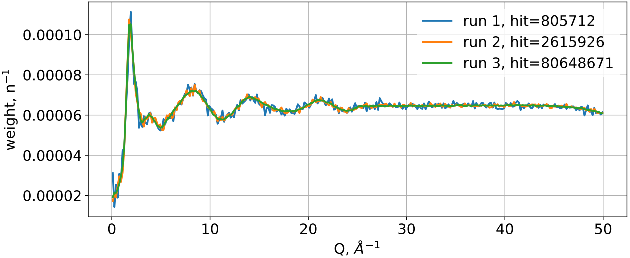
- incident monoenergetic $0.15\,\text{Å}$ neutron
- CAB heavy water spherical sample
- a hypothetical energy and position sensitive 4π detector



Neutron weighted structure factor

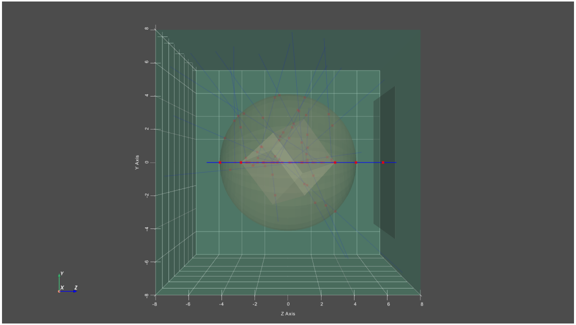
run	neutron	HW bias	time(s)
1	1e7	1.0	15
2	1e7	8.0	25
3	1e9	1.0	1461

Three runs measuring the distribution of Q for different number of scatterings. The advanced variance reduction technique is tested.



Optimisation (experimental feature in V2)

Finding the optimal position of a collimated point source (based on Optuna `optuna.org`)

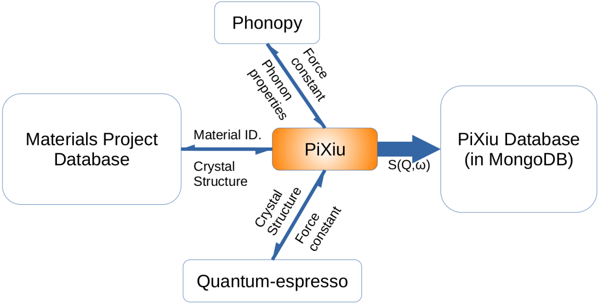


1. Objective: the count rate of the detector is minimal.
2. Variables: the point source position.
3. Constraint: the position is at the surface of the sphere.

Results: optimised position

- The optimal position is expected at [0,0,-5]
- In 100 iterations, the best trial position is [-1.31, 1.09, -4.85]
- In 1000 iterations, the best trial position is [0.10, 0.16, -5.00]

PiXiu(貔貅)

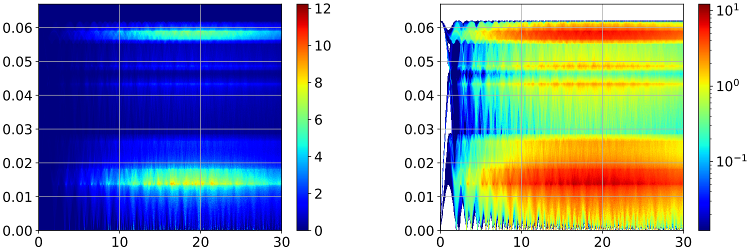


- PiXiu is capable of calculating the scattering function, $S(\vec{Q}, \omega)$, in four dimensions. A powder-averaged $S(Q, \omega)$ is generated by default.
- PiXiu package utilises the Apache Spark engine for parallelisation and high-throughput computation.

PiXiu for direct inelastic instruments

An example of calculating silicon:

```
px_pre.py --mp-id mp-149 --numcpu 64 --rundft
px_inelastic_direct.py --temperature 30 --upper-limit-Q 30
```

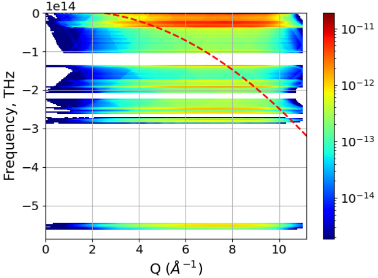


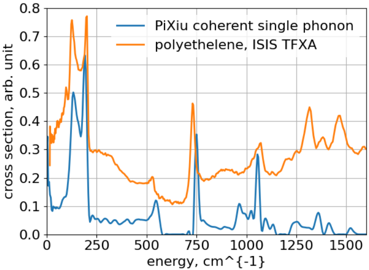
The program occupies 64 cores and used 20mins to complete. The resolution is 0.05/Aa over Q and 0.15meV over energy.

PiXiu for indirect inelastic instruments

An example of calculating polyethylene:

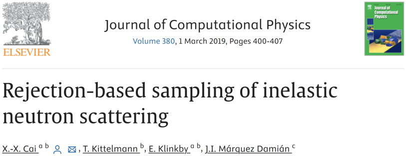
```
px_pre.py --mp-id mp-985782 --vanDerWaals --numcpu 256 --rundft
px_inelastic_direct.py --temperature 20 --upper-limit-Q 10
px_inelastic_indirect.py --scattering-angle 135 --energy-out 0.00384
```





Sampling in isotropic materials

It is considered as a solved problem.



ELSEVIER

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Rejection-based sampling of inelastic neutron scattering

X.-X. Cai [a] [b], T. Kittelmann [b], E. Klinkby [a] [b], J.I. Márquez Damián [c]

- Overcoming artefacts introduced by conventional interpolation methods.
- Reproducing continuous energy and angular spectra with high numerical accuracy.
- Close to unity acceptance rate in general cases.
- Straightforward for an existing code to adapt.

Double differential and integral cross sections

The double differential cross section is

$$\frac{\partial^2\sigma}{\partial E'\partial\Omega} = \frac{\sigma_b}{4\pi}\frac{k'}{k}S(Q,\omega) = \frac{\sigma_b}{4\pi}\sqrt{\frac{E'}{E}}\frac{S(\alpha,\beta)}{k_b T} \tag{1}$$

with

$$\alpha = \frac{E + E' - 2\mu\sqrt{E'E}}{k_b T} \quad \text{and} \quad \beta = \frac{E' - E}{k_b T} \tag{2}$$

The integral cross section is

$$\sigma(E) = \frac{C}{E}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} S(\alpha,\beta)\,\Theta(\alpha,\beta)\,\mathrm{d}\alpha\,\mathrm{d}\beta \tag{3}$$

where C is a constant that equals $\sigma_b k_b T/4$, and

$$\Theta(\alpha,\beta) = \begin{cases} 1 & \text{when} \quad (\alpha,\beta)\in D \\ 0 & \text{otherwise} \end{cases} \tag{4}$$

Rejection sampling

The distribution of scattered neutron can be found as

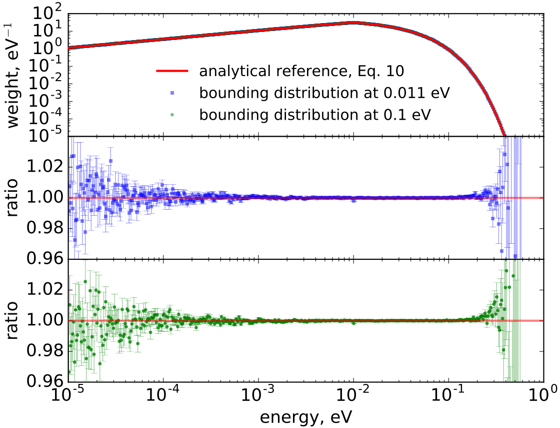
$$p(\alpha, \beta\,|E_l) = \frac{\sigma(E_h)E_h}{\sigma(E_l)E_l} p(\alpha, \beta\,|E_h)\,\Theta_l(\alpha, \beta) \tag{5}$$

The corresponding numerical steps are as followed

```
1  find smallest i, so that $E \leq E_i$
2  repeat
3      sample a $\beta'$ from $P(\beta\,|\,E_i)$
4  until $-E/k_b T \leq \beta$;
5  find j, so that $\beta_{j-1} \leq \beta' \leq \beta_j$
6  sample $\alpha_l$ from $F(\alpha\,|\,\beta_{j-1}, E_i)$
7  sample $\alpha_h$ from $F(\alpha\,|\,\beta_j, E_i)$
8  interpolate an $\alpha'$ from $\alpha_l$ and $\alpha_h$
9  if $\alpha' \notin (\alpha_-, \alpha_+)$ then go to 2 ;
10 else accept $\alpha'$ and $\beta'$ ;
```

Benchmark

Comparisons between the distribution of free gas analytic expression and those sampled from two different cross section tables. The numerical performance is robust and independent of input data energy range.



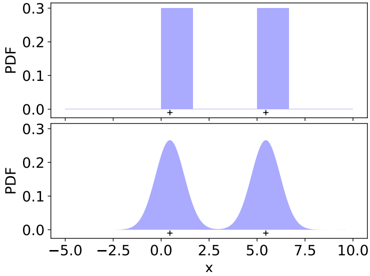
Sampling in single crystals

- The rejection method is proven to be robust and accurate when applies to isotropic material.
- It is challenging to implement that for the 4D scattering function, i.e. $S(\vec{\mathrm{Q}}, \omega)$, in the step 3, 6 and 7 of the algorithm.
  1. The memory footprint of the data table grows exponentially with neutron energy and distribution resolution.
  2. The implementation is numerical cumbersome. It is difficult to have robustness and accuracy at the same time.

It has been found that kernel density estimation is a better alternative for the sampling.

Kernel density estimation

Kernel density estimation (KDE) can also represent an unknown distribution. Each event is broaden by a kernel function that is straight forward to manipulate in high dimension.



Example representations of two events from an 1D distribution

In KDE, a kernel represents a phonon with a finite line shape. It is physically sound, as phonon is a type of quasiparticle.

Validation

Numerical experiments are performed to test the idea.

- Use the histogram method to calculate the 2D power pattern of silicon, as the reference.
- Use KDE to sample 10 million scatterings with random crystal orientation, and analyse for the power pattern.
- The obtain patterns are in principle identical.

Numerical procedure

The procedures are as followed

1. Generate 1e6 points in a cubic region of the Q space.
2. Use PiXiu to calculate phonon scattering cross sections of each branch.
3. Use KDSource[1] to classify phonons and create the parameters of the KDE object.
4. Sample 1e7 scattering events from the parameters.
5. Analysis the scattering events to create a powder pattern.

It has been found that a single core completes the step 4 in 4.8s. That indicates the practicality of using this method in Monte Carlo simulations.

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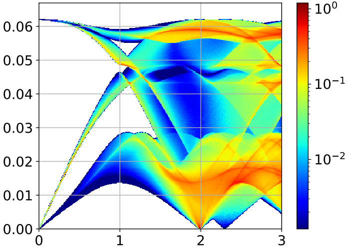
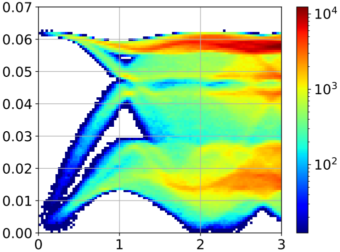
[1] available at `github.com/KDSource`

Results

Reference from PiXiu on the left, sampled result on the right.





- The results are broadly similar.
- The new sampling method failed to capture the contributions near the gamma points.
- The resolution of the sampled result is not optimal.

Outlook

- The reference paper of PiXiu will be made available on arXiv very soon. The source code will be released under an open source license, like all our other codes.
- It is in evident that the new sampling method is practical.
- Unlike PiXiu, the current method does not know any important region in the Q space. New algorithm is under development of producing data points in importance regions to train the KDE.