# NEW SCATTERING KERNELS FOR SOLID DEUTERIUM AND SUPERFLUID He-4

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PART 1

# A New Scattering Kernel for Solid $D_2$

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The scattering law of a molecular system is

$$S(\mathbf{Q},\omega) = \frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} dt e^{-i\omega t} \left\langle \sum_{l,l'} \sum_{\nu,\nu'} \overline{a^*_{l\nu} a_{l'\nu'}} \exp\{-i\mathbf{Q}.\mathbf{R}_{l\nu}(0)\} \exp\{i\mathbf{Q}.\mathbf{R}_{l'\nu'}(t)\} \right\rangle$$

where $\mathbf{R}_{l\nu}(t)$ denotes the position of the atom $\nu$ within the molecule $l$ ,

$$\mathbf{R}_{l\nu}(t) = \mathbf{a}_l + \mathbf{b}_\nu(t) + \mathbf{u}_\nu(t)$$

and can be written as the sum of inter ($l \neq l'$)- and intra ($l = l'$)- molecular contributions (also referred to as the *outer* and *inner* terms, respectively).

The outer and inner terms, for the intermediate scattering function are

$$\chi(\mathbf{Q},t) = \left\langle \sum_{l \neq l'} \sum_{\nu,\nu'} \overline{a_{l\nu}^{*} a_{l'\nu'}} \exp\{-i\mathbf{Q}.\mathbf{R}_{l\nu}(0)\} \exp\{i\mathbf{Q}.\mathbf{R}_{l'\nu'}(t)\} \right\rangle + \qquad \text{"Outer"}$$

$$\left\langle \sum_{l} \sum_{\nu,\nu'} \overline{a_{l\nu}^{*} a_{l\nu'}} \exp\{-i\mathbf{Q}.\mathbf{R}_{l\nu}(0)\} \exp\{i\mathbf{Q}.\mathbf{R}_{l\nu'}(t)\} \right\rangle \qquad \text{"Inner"}$$

Here the brackets denote the average of the time-dependent operators over an equilibrium-distribution function in the full phase space of the scattering system.

In terms of the usual coherent, $b_c^{\nu}$ , and incoherent, $b_i^{\nu}$ , scattering lengths for nuclei $\nu$,

$$a_{\nu} = b_c^{\nu} + 2b_i^{\nu}(\mathbf{S}_{\nu}.\mathbf{s})\left[S_{\nu}(S_{\nu}+1)\right]^{-1/2}$$

Leaving aside for the moment the consideration of vibrational modes:

$$\chi^{out}(\mathbf{Q},t) = \underbrace{\sum_{l\neq l'} \left\langle \exp\{-i\mathbf{Q}.\mathbf{a}_l(0)\}\exp\{i\mathbf{Q}.\mathbf{a}_{l'}(t)\}\right\rangle}_{I_d(\mathbf{Q},t)} . \underbrace{\sum_{v,v'} \left\langle \overline{b_{lv}^*}\exp\{-i\mathbf{Q}.\mathbf{b}_{lv}(0)\}\overline{b_{l'v'}}\exp\{i\mathbf{Q}.\mathbf{b}_{l'v'}(t)\}\right\rangle}_{u(\mathbf{Q},t)}$$

$$\therefore \quad \chi^{out}(\mathbf{Q},t) = 4\,(b_c)^2\, j_0^2\,(Qd/2)\,.\, I_d(\mathbf{Q},t)$$

$$\chi^{inner}(\mathbf{Q},t) = \underbrace{\sum_{l} \left\langle \exp\{-i\mathbf{Q}.\mathbf{a}_l(0)\}\exp\{i\mathbf{Q}.\mathbf{a}_{l}(t)\}\right\rangle}_{I_s(\mathbf{Q},t)} . \underbrace{\sum_{JJ'} \sum_{\pi\pi'} A_{\pi\pi'}(J)\, f_{\pi\pi'}(Q;J,J')\, e^{-i(\omega_J-\omega_{J'})t}}_{v(\mathbf{Q},t)}$$

$$\therefore \quad \chi^{inner}(\mathbf{Q},t) = v(\mathbf{Q},t)\,.\, I_s(\mathbf{Q},t)$$

$$\boxed{\chi(\mathbf{Q},t) = 4\, b_c^2\, j_0^2\, (Qr/2)\, \{I\, (\mathbf{Q},t) - I_s\, (\mathbf{Q},t)\} + v(\mathbf{Q},t)\, .\, I_s\, (\mathbf{Q},t)}$$

Now, (must multiply everything by $\chi^{vib}(\mathbf{Q},t)$ !)

$$\chi(\mathbf{Q},t) = \underset{\text{(elast)}}{\chi(\mathbf{Q},0)} + \underset{\text{(inelast)}}{\chi(\mathbf{Q},t\neq 0)}$$

but because

$$I_s\, (\mathbf{Q},0) = 1 \qquad , \qquad I(\mathbf{Q},0) = |F(\mathbf{Q},0)|^2$$

$$\chi^{el}(\mathbf{Q},0) = 4\, b_c^2\, j_0^2\, (Qr/2)\, |F(\mathbf{Q})|^2 + v(\mathbf{Q},0) - u(\mathbf{Q})$$

within the Incoherent Approximation: $\boxed{I(\mathbf{Q},t \neq 0) \cong I_s(\mathbf{Q},t \neq 0)}$

$$\chi^{inel}(\mathbf{Q},t) = v(\mathbf{Q},t)\, .\, I_s\, (\mathbf{Q},t)$$

# OLD MODEL

The old calculations were performed according to:

$$\chi^{el}(\mathbf{Q},0) = 4\, b_c^2\, j_0^2\, (Qr/2)\, |F(\mathbf{Q})|^2\, \chi^{vib}(\mathbf{Q},0) \qquad \text{Convent.El.Coh}$$

$$+ 2\,(1+\alpha)\, b_i^2\, \chi^{vib}(\mathbf{Q},0) \qquad \text{Total El.Incoh}$$

($\alpha$ = ¼ for o-D2, - ½ for p-D2, 0 for n-D2)

$$\chi^{inel}(\mathbf{Q},t) = v(\mathbf{Q},t)\; .\; I_s\,(\mathbf{Q},t)\; .\; \chi^{vib}(\mathbf{Q},t) \qquad \text{Total Inelast.}$$

# NEW MODEL

Convent.El.Coh

Convent.El.Incoh

$$S^{el}(\mathbf{Q},0) = \left[4\, b_c^2\, j_0^2\, (Qd/2)\, |F(\mathbf{Q})|^2 + 2\, b_i^2 + \right.$$

$$\left. + 2\, b_i^2\, \alpha\, j_0\, (Qd) + 2\, b_c^2\, \{1 + j_0\, (Qd) - 2\, j_0^2\, (Qd/2)\}\right] e^{-2W}$$

Spin Correlation

Δ structure

$$S^{inel}(\mathbf{Q},\omega) = \left[4\, b_c^2\, j_0^2\, (Qd/2)\, \{S^{1ph}(\mathbf{Q},\omega) - S_s^{\,1ph}(\mathbf{Q},\omega)\} + V_o^{rot}(\mathbf{Q},\omega) \otimes S_s(\mathbf{Q},\omega)\right] e^{-2W}$$

Correction to 1ph IA

Convent.Inel.Incoh

**Coherent Neutron Scattering in Polycrystalline Deuterium and its Implications for Ultracold Neutron Production**

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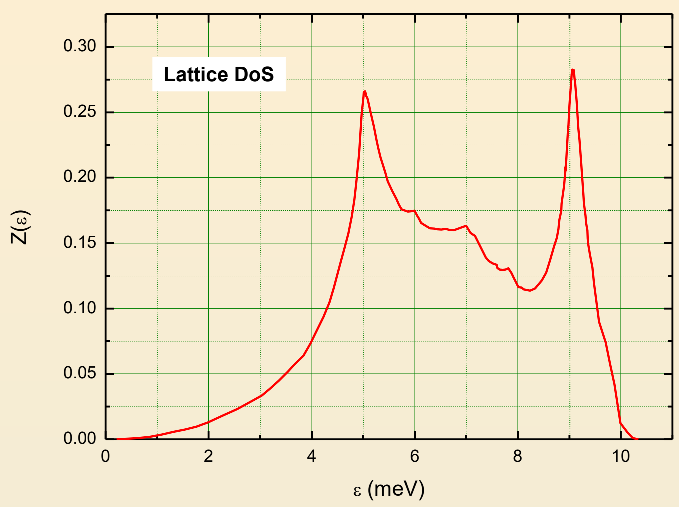
*2. Physics Department, North Carolina State University, Raleigh, NC 27695*
(Dated: May 7, 2010)

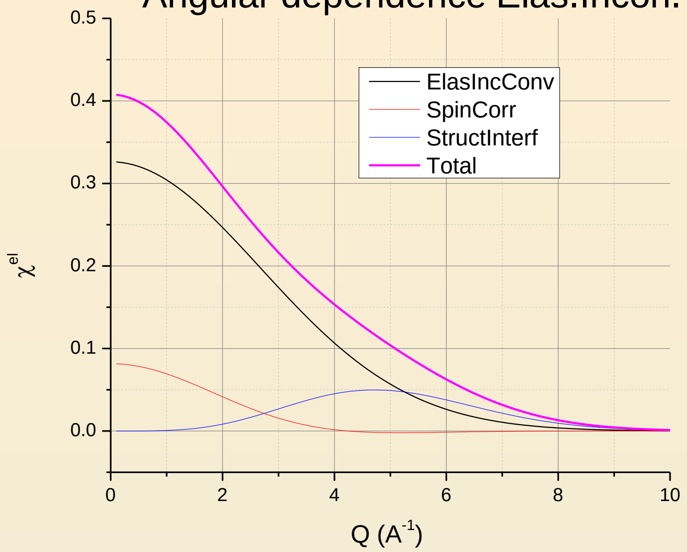
Marked terms: not included in standard NJOY´s algorithm.

Sep2016

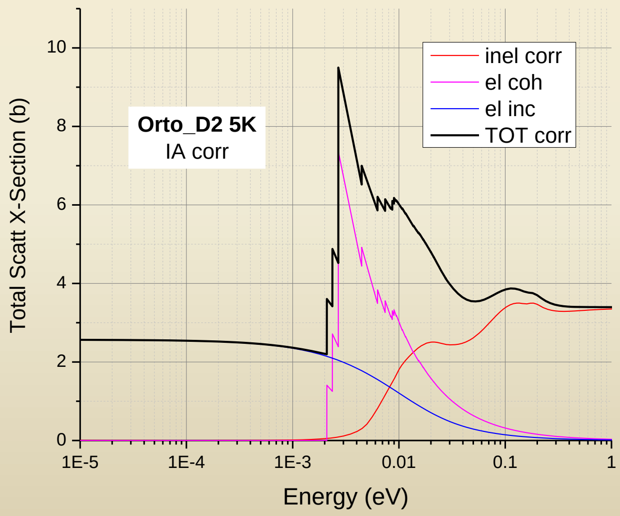
## Phonon Density of states

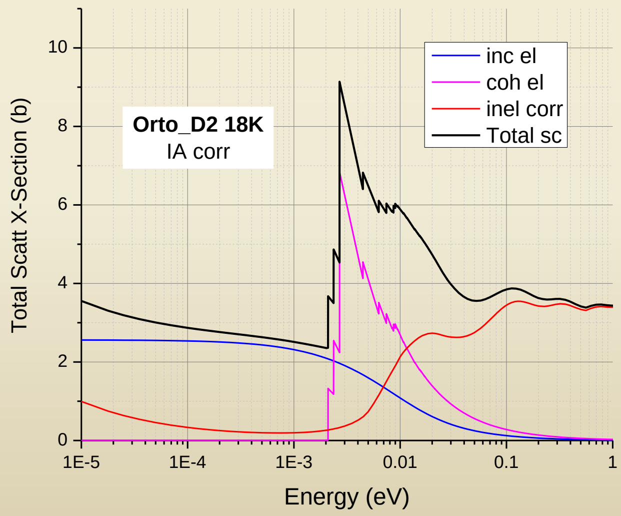



## Angular dependence Elas.Incoh.




## Cross sections per atom



## CORRECTION TO THE ONE-PHONON INCOH. APPROX.

At cold and very cold neutron energies, the total inelastic cross section for $sD_2$ at low temperatures will be dominated by the lattice one-phonon upscattering processes, as there are no rotational nor vibrational excitations thermally or collisionally able to participate. We obtain, for *ortho*-deuterium under those conditions:

$$S^{inel(1)}(\mathbf{Q},\omega) = [4\, b_c^2\, j_0^2\,(Qr/2)\,\{S^{1ph}(\mathbf{Q},\omega) - S_s^{\ 1ph}(\mathbf{Q},\omega)\} +$$

$$+ \{2\,(b_c^2 + b_i^2) + (2b_c^2 + \tfrac{1}{2}\, b_i^2)\, j_0(Qd)\}\, S_s^{\ 1ph}(\mathbf{Q},\omega)]\, e^{-2W}$$

and in the limit of very small energy and momentum transfer:

$$S^{inel(1)}(\{\mathbf{Q},\omega\}\rightarrow 0) = 4\, b_c^2\, S^{1ph}(\mathbf{Q},\omega) + 5/2\, b_i^2\, S_s^{\ 1ph}(\mathbf{Q},\omega)$$

The application of the IA in these Eqs. means replacing $S^{1ph}(\mathbf{Q},\omega)$ by $S_s^{\ 1ph}(\mathbf{Q},\omega)$.

It is convenient to define the ratio of the coherent and incoherent one-phonon scattering functions:

$$\delta S^1(\mathbf{Q},\omega) = S^{1ph}(\mathbf{Q},\omega) - S_s^{\ 1ph}(\mathbf{Q},\omega)$$

The ratio $\{\delta S^1 / S_s^{\ 1ph}\}_T$ derived from the exact calculations for solid o-$D_2$ at 0.33 μeV as a function of temperature (Liu *et al*) can be well approximated by

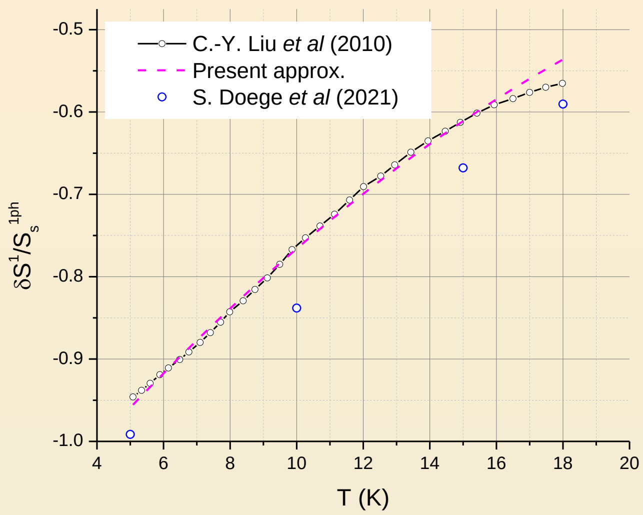
$$\{\delta S^1 / S_s^{\ 1ph}\}_T \approx -1.19 + 5\ 10^{-2}\ T(K) - 7.3\ 10^{-4}\ T(K)^2\ , \quad T \leq 16K$$

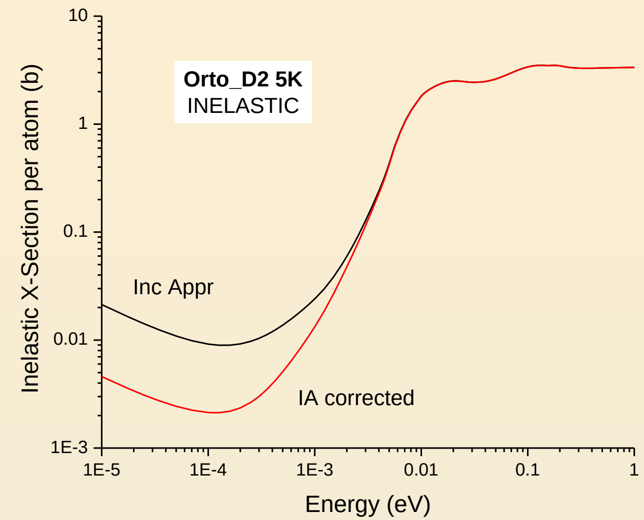
Besides the exact procedure that involves a MC sampling of the dispersion relations over the appropriate phase space to evaluate the coherent one-phonon term $S^{1ph}(\mathbf{Q},\omega)$, it seems plausible to approximate

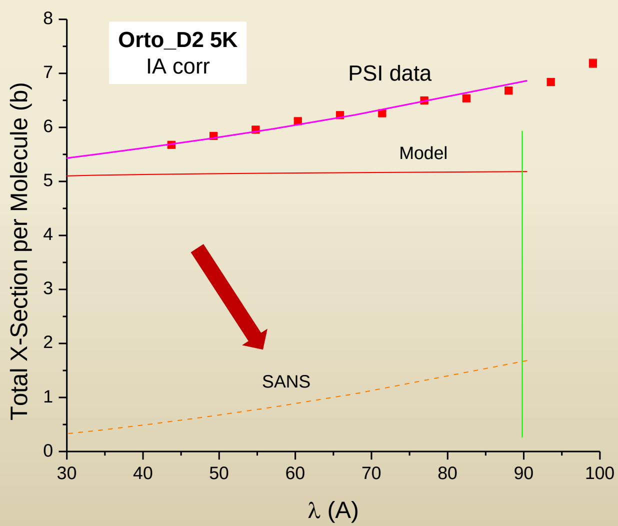
$$\delta S^1(\mathbf{Q},\omega) = S^{1ph}(\mathbf{Q},\omega) - S_s^{\ 1ph}(\mathbf{Q},\omega) \approx \{\delta S^1 / S_s^{\ 1ph}\}_T\ .\ S_s^{\ 1ph}(\mathbf{Q},\omega)$$

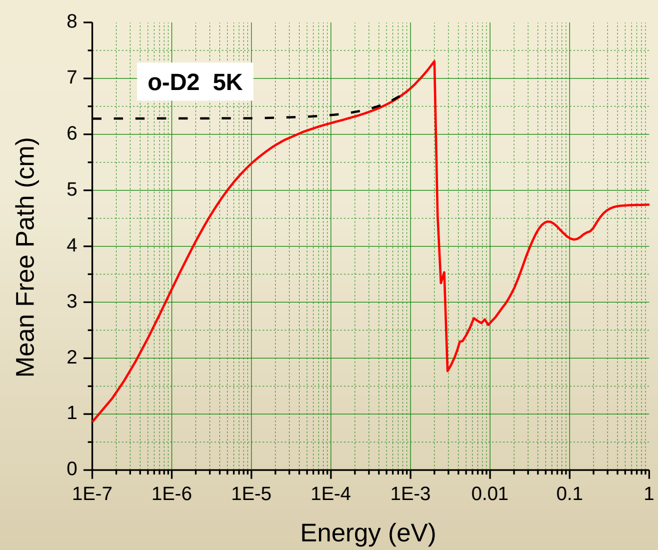
so that

$$S^{inel}(\mathbf{Q},\omega) = [4\ b_c^2\ j_0^2\ (Qr/2)\ \{\delta S^1 / S_s^{\ 1ph}\}_T\ .\ S_s^{\ 1ph}(\mathbf{Q},\omega) + V_o^{rot}(\mathbf{Q},\omega) \otimes S_s(\mathbf{Q},\omega)\ ]\ e^{-2W}$$


C.-Y. Liu et al (2010)
Present approx.
S. Doege et al (2021)
δS1/Ss1ph
T (K)


Orto_D2 5K
INELASTIC
Inc Appr
IA corrected
Inelastic X-Section per atom (b)
Energy (eV)


Orto_D2 5K
IA corr
PSI data
Model
SANS
Total X-Section per Molecule (b)
λ (A)


o-D2 5K
Mean Free Path (cm)
Energy (eV)

PART 2

# A New Scattering Kernel for Superfluid $^4$He

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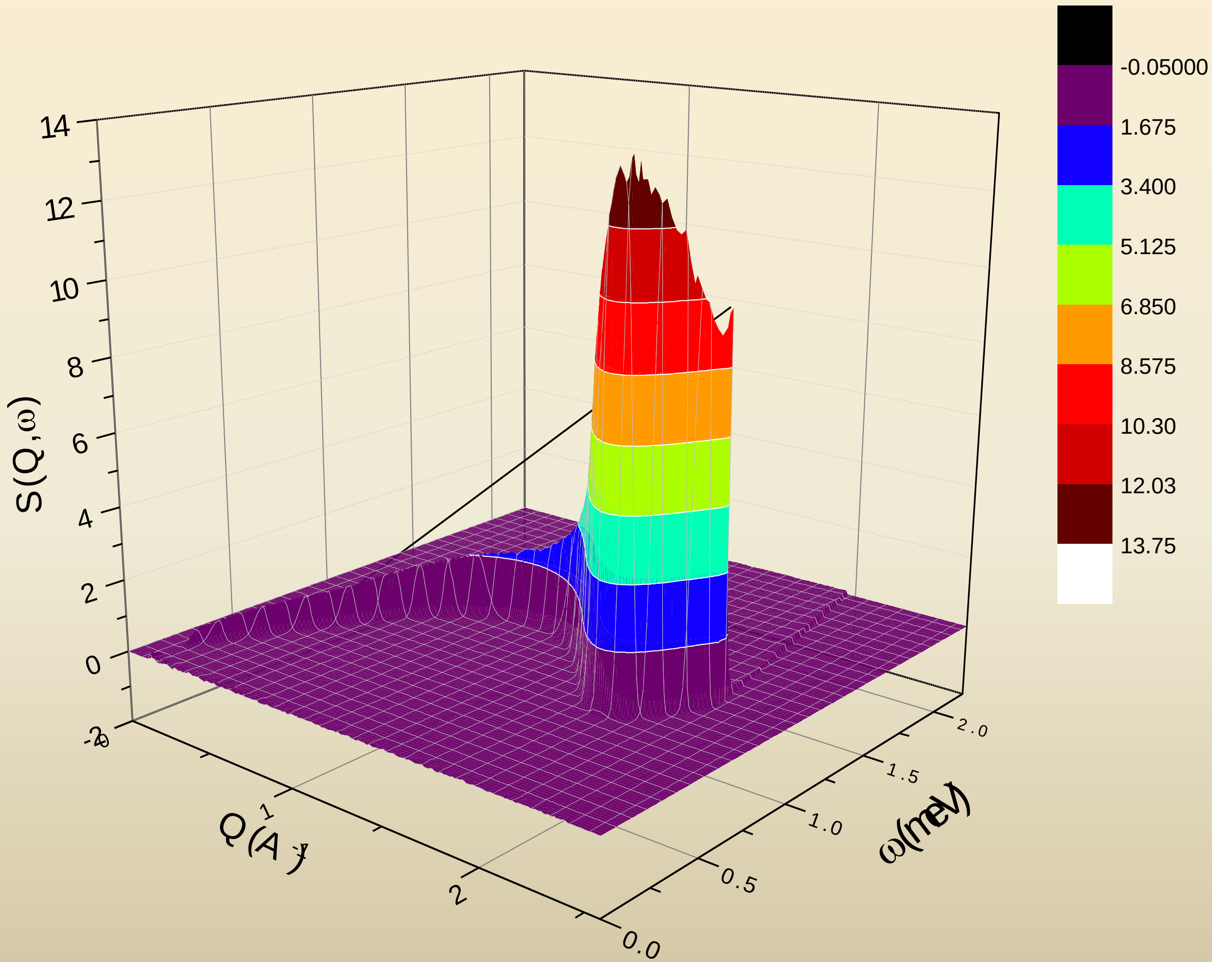
PHYSICAL REVIEW B **103**, 104516 (2021)

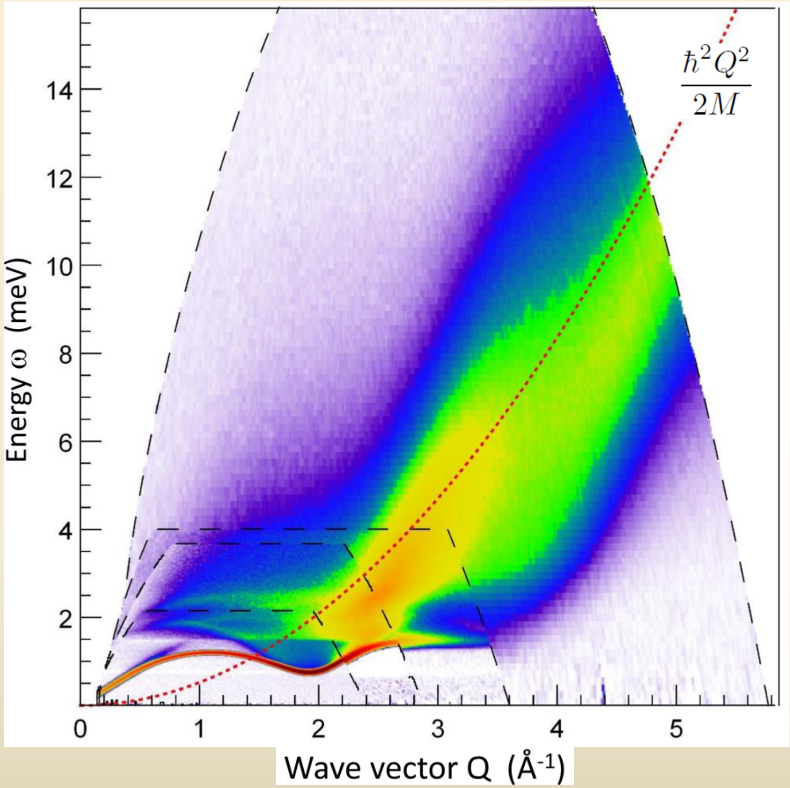
Editors' Suggestion    Featured in Physics

# Dispersion relation of Landau elementary excitations and thermodynamic properties of superfluid $^4$He

H. Godfrin,[1,*] K. Beauvois,[1,2] A. Sultan,[1,2] E. Krotscheck,[3,4] J. Dawidowski,[5] B. Fåk,[2] and J. Ollivier[2]



At low momentum transfer and temperatures $\lesssim$ 1 K, neutron scattering at low energies from superfluid $^4$He is dominated by the collective phonon-roton mode.



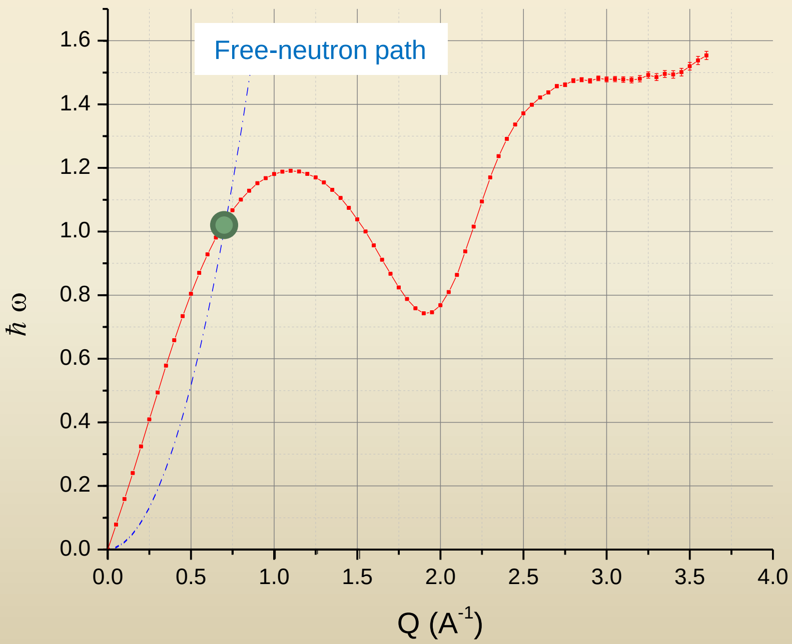

*S(Q,ω) of superfluid 4He as a function of wave vector and energy transfer, measured at T ≲100 mK*

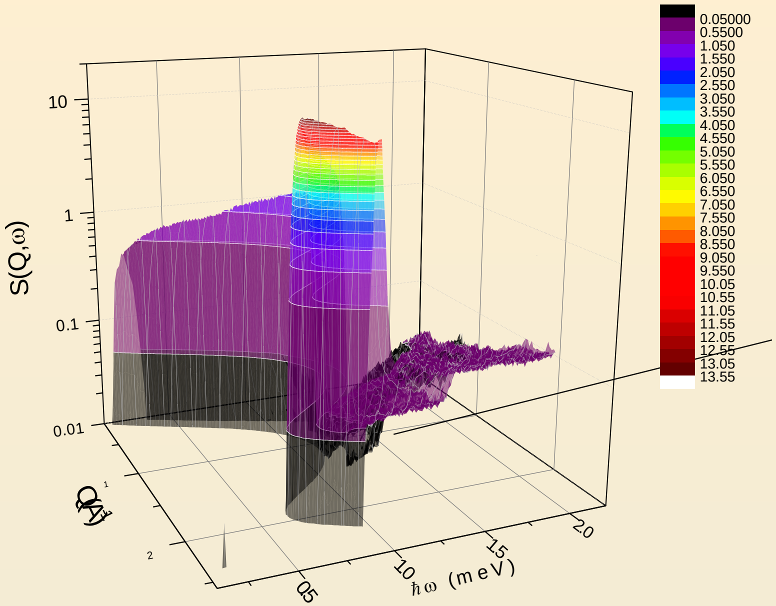
K. Beauvois *et al., Phys.Rev.* B **97**, 184520 (2018)

This unique property makes superfluid $^4$He, in addition to it's zero absorption cross section and limited up-scattering due to the low temperature, an attractive option as a source of ultra-cold neutrons (UCNs) at neutron scattering facilities.

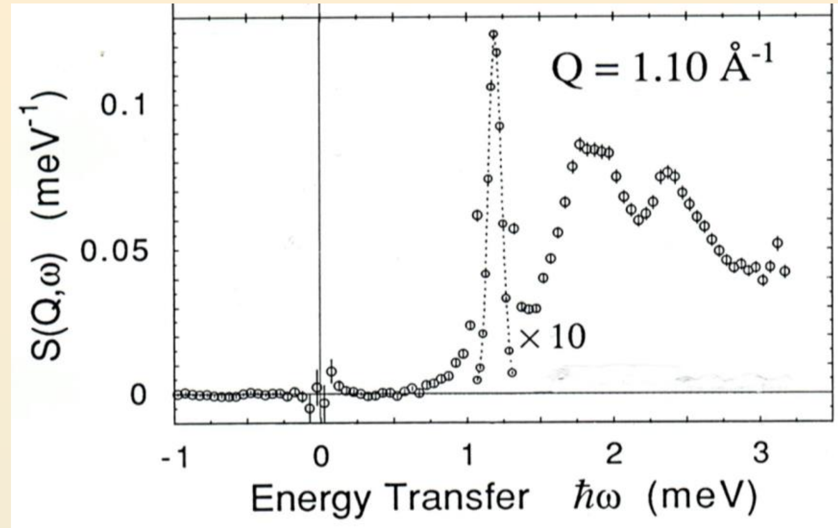
*Dispersion relation $\varepsilon$ (Q) of the single excitations*

The primary mechanism for the production of UCNs is characterized by a downscattering process at the crossing point between a free neutron and the phonon-roton dispersion curve, which happens around a neutron energy of 1meV.






H. Godfrin (priv. comm.)




K. Andersen *et al.* (1994)

At temperatures below T ≈ 1.3 K, the neutron scattering from superfluid $^4$He can be readily separated into clear contributions due to single- and multi-phonon excitations, represented by

$$\boxed{S(Q,\omega) = S_s(Q,\omega) + S_m(Q,\omega)}$$

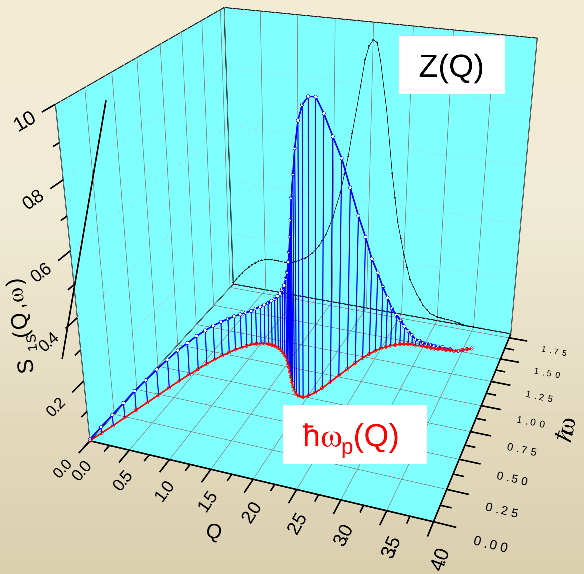
where $S_S(Q, \omega)$ and and $S_M(Q, \omega)$ are the scattering functions for single-phonon excitations and for multi-phonon excitations, respectively(*)

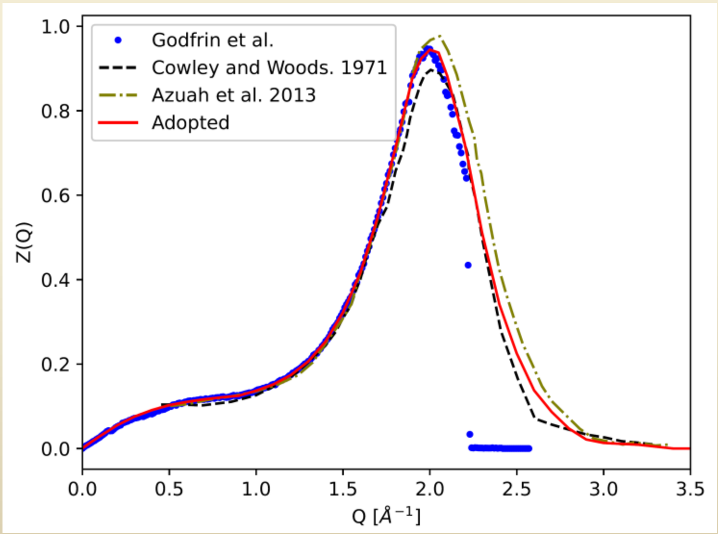
(*) A. Miller, D. Pines, P. Noziéres, Phys.Rev. **127**, 1452 (1962)

For the single-excitation term in this range we have used a Lorentzian of the form

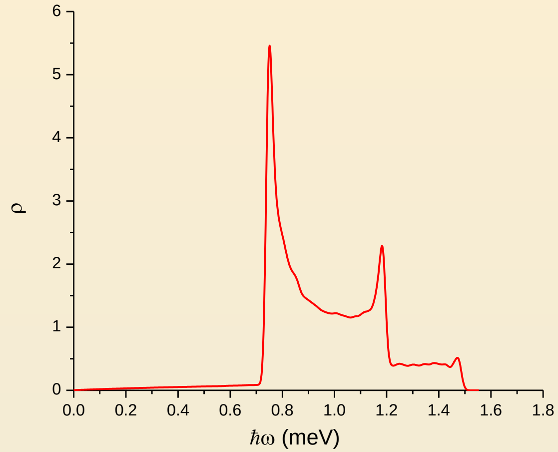
$$S_s(Q,\omega) = \frac{Z(Q)}{\pi}\frac{\Gamma(Q)}{(\hbar\omega - \hbar\omega_p(Q))^2 + \Gamma(Q)^2}$$

where $\hbar\omega_p(Q)$ is the energy of the single-phonon excitations and $\Gamma(Q)$ is the halfwidth at half maximum and depends on the temperature of the liquid. *Z(Q)* is the single-phonon structure factor that measures its intensity.



By denoting $S_s^{exp}(\omega)$ as the integral over Q of the experimental dispersion curve, $S_s^{exp}(Q,\omega)$, the phonon weighted frequency distribution can be calculated by




$$\rho(\omega) = \frac{\hbar\omega}{k_B T}[e^{\frac{\hbar\omega}{kT}} - 1] S_s^{\exp}(\omega)$$

The multiphonon term of the total scattering function, applying the Sköld approximation, is then

$$S_M(Q,\omega) = H(Q) S_M^{\mathrm{self}}\left(\frac{Q}{\sqrt{u(Q)H(Q)}}, \omega\right)$$

With this definition, and the form of $S_s(Q,\omega)$, the total scattering function $S(Q,\omega)$ will satisfy the first Sum Rule

$$S(Q) = \int_{-\infty}^{\infty} S(Q,\omega)d\omega = Z(Q) + H(Q)$$

As long as H(Q) /

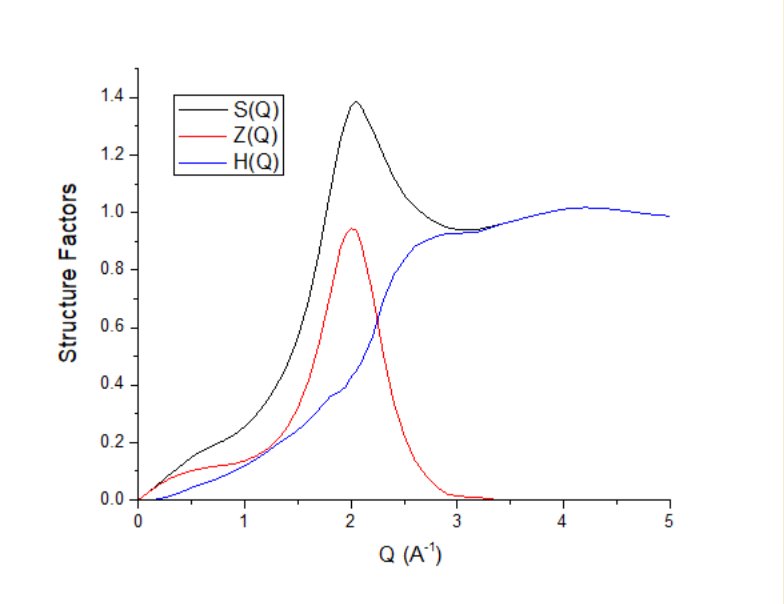
$$H(Q) = S(Q) - Z(Q)$$

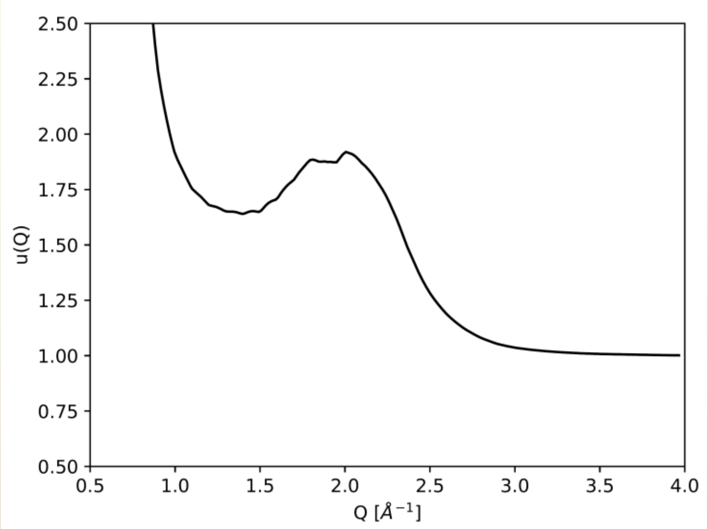
where both structure factors, S(Q) and Z(Q), are measured quantities.
Using the value v = 238.3m/s, we obtain the slope of S(Q → 0) at very low temperatures.

To satisfy the second Sum Rule

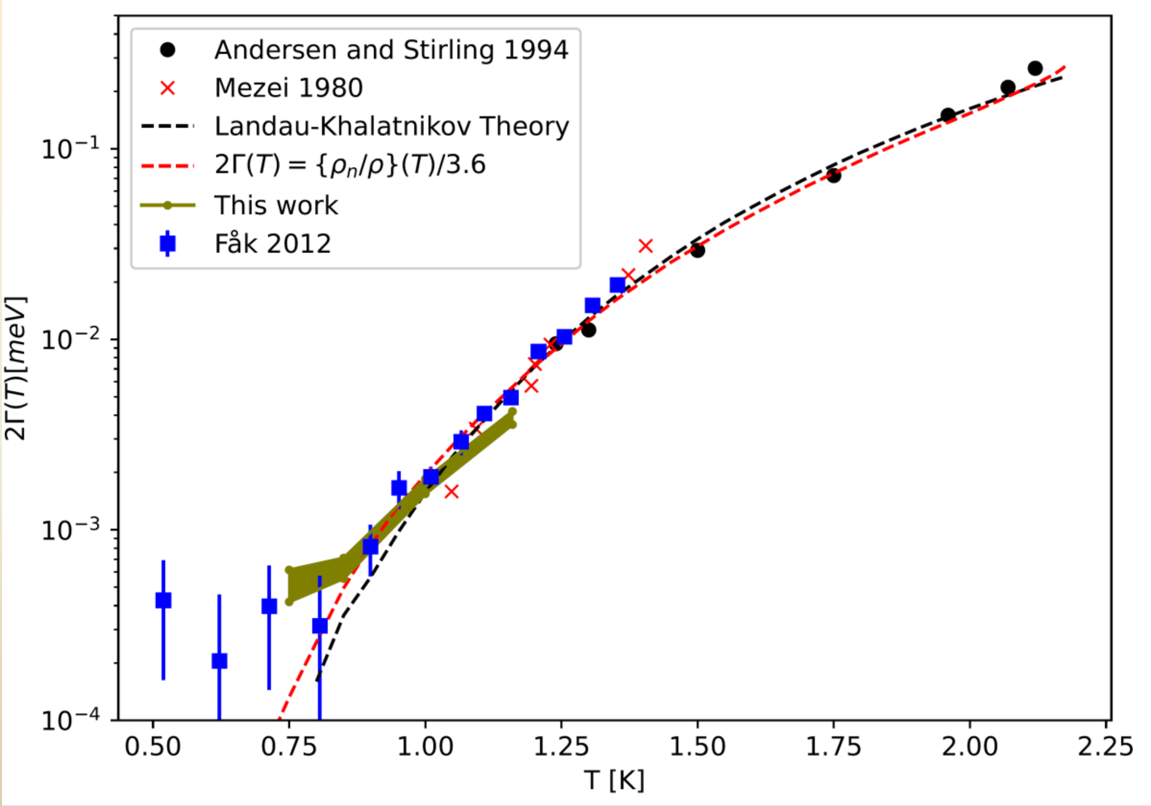
$$\boxed{\frac{\hbar^2 Q^2}{2M} = \hbar \int_{-\infty}^{\infty} \omega S(Q,\omega) d\omega}$$

an effective mass Mu(Q) has been introduced in the expression of $S_M(Q, \omega)$.



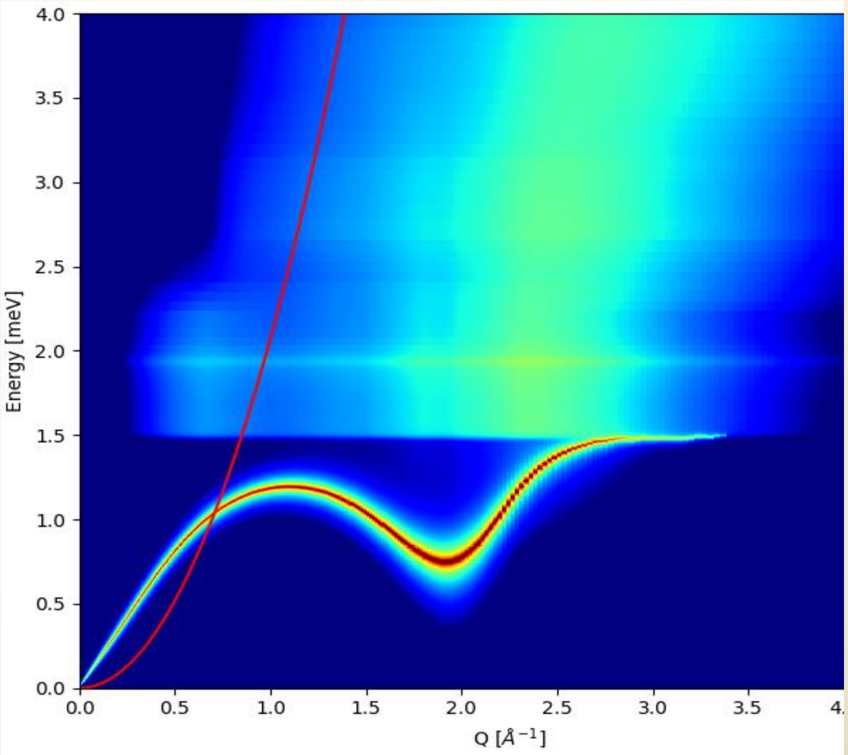



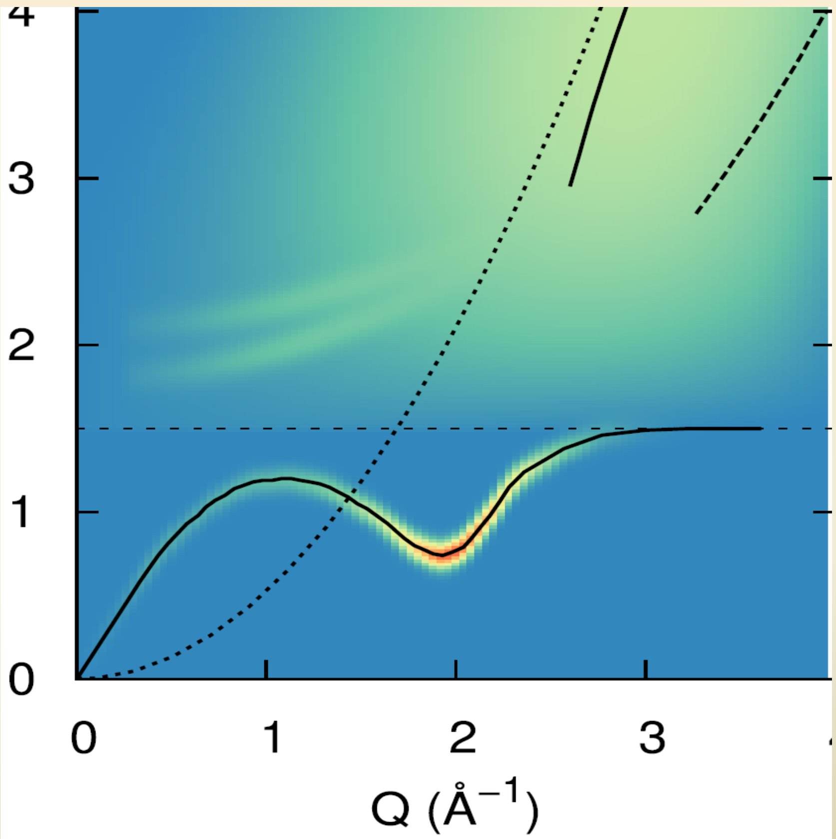
The formalism has been coded into a custom version of the LEAPR module of NJOY2016 . The input to the calculations include the phonon-roton dispersion curve, the structure factors, the weighted frequency distribution, the effective mass function, and a normalizing function for the incomplete phonon expansion, which is calculated internally.



# THE SCATTERING FUNCTION S(Q, $\omega$)

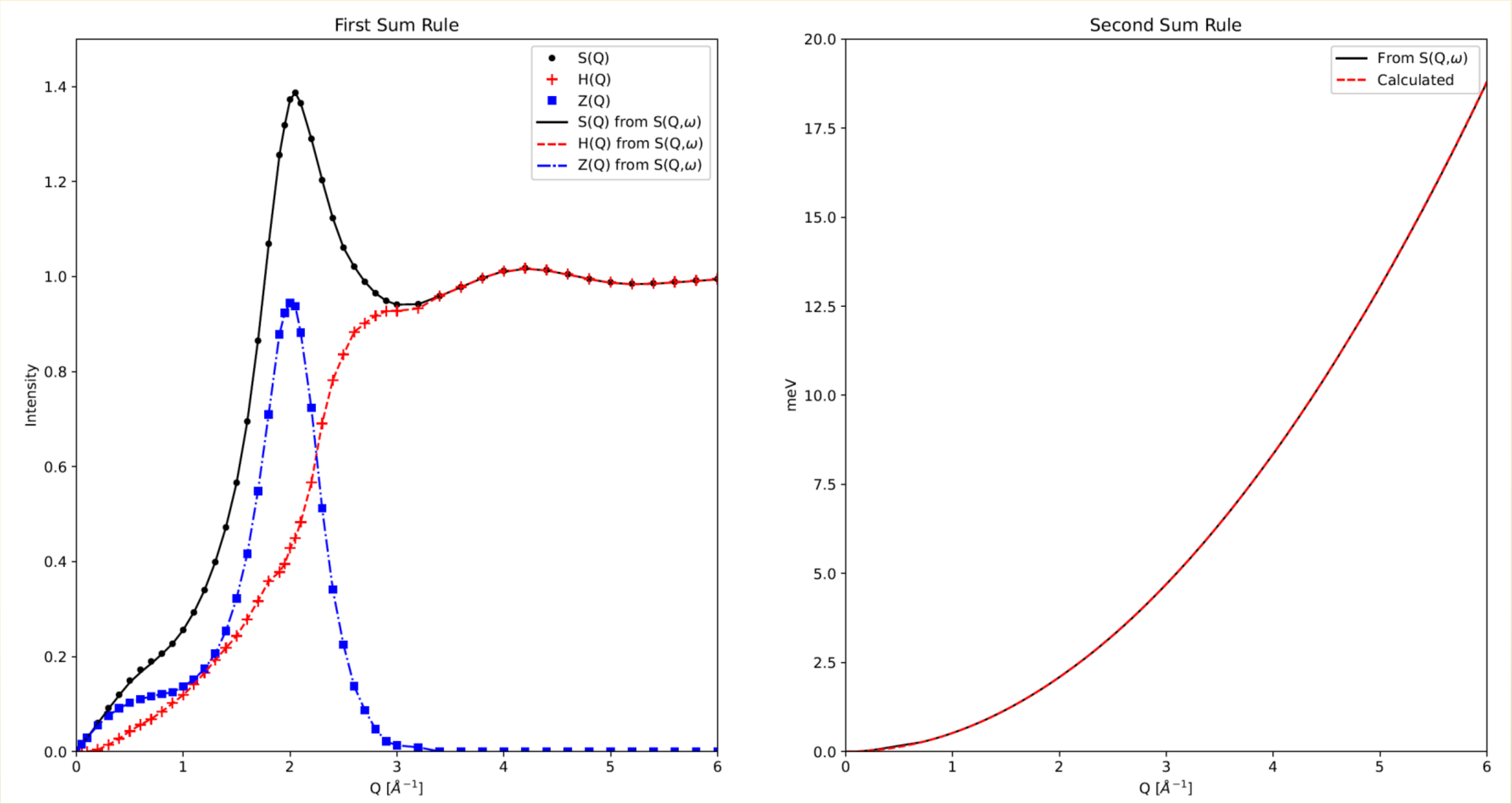
Calculated scattering kernel at 1.3 K. The red line is the dispersion curve of a free neutron.



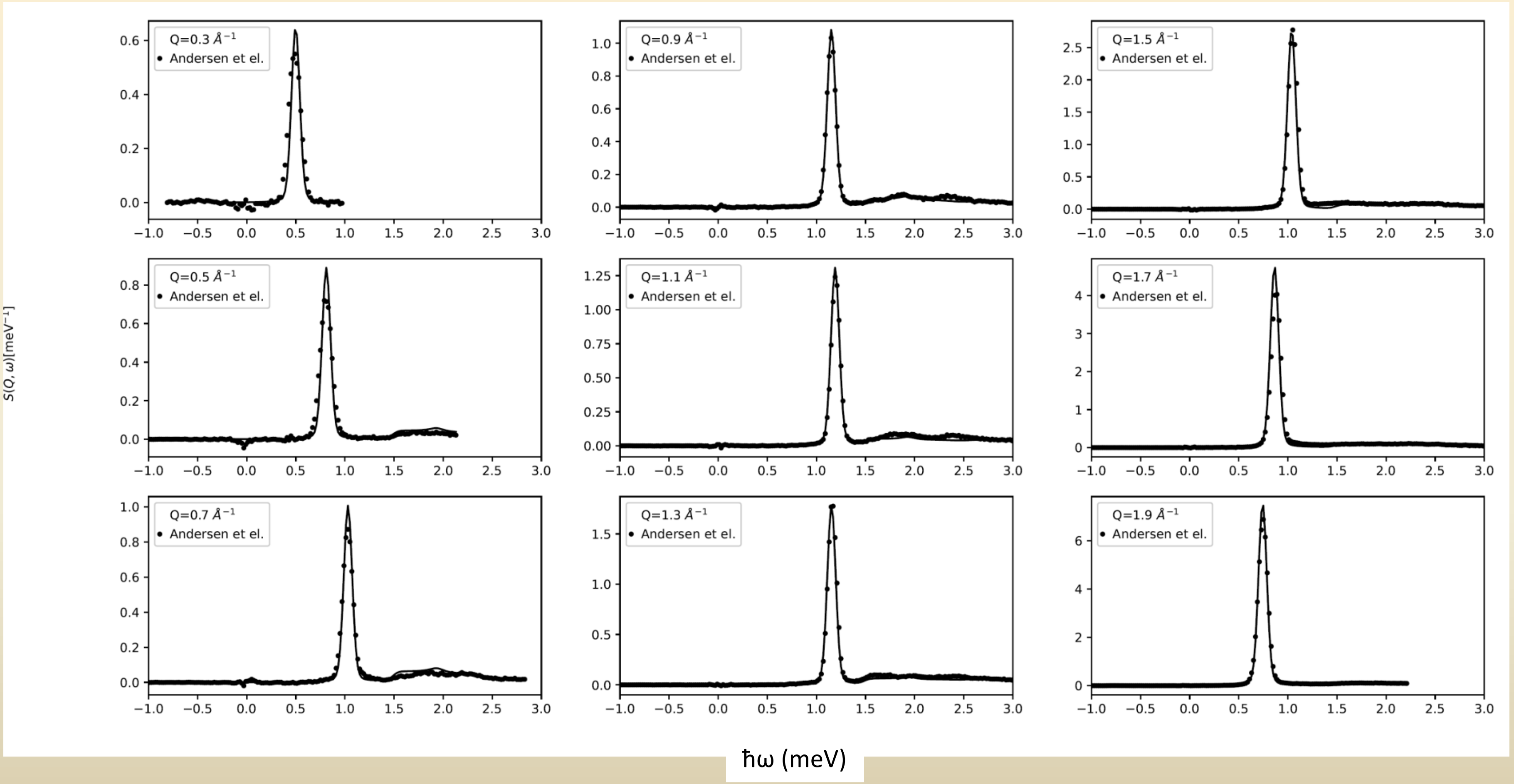

*Adapted from Glyde (2018)*

# SUM RULES



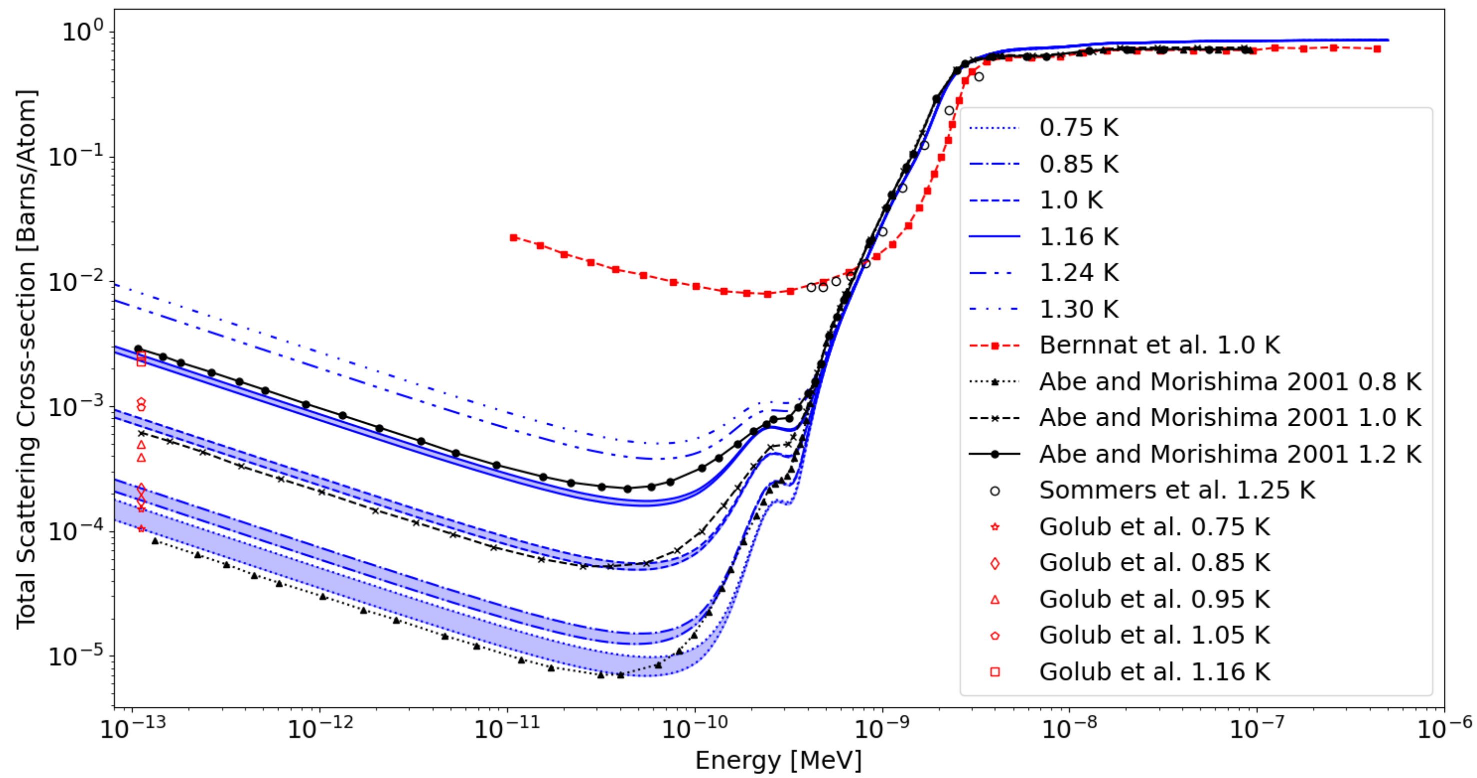

Projected structure factors obtained from calculations at 1.3 K, compared to the input data (left). Calculated second sum rule compared to the second sum rule calculated from the scattering kernel at 1.3K (right).

Dynamic structure factor at 1.3K convoluted with an instrument resolution function

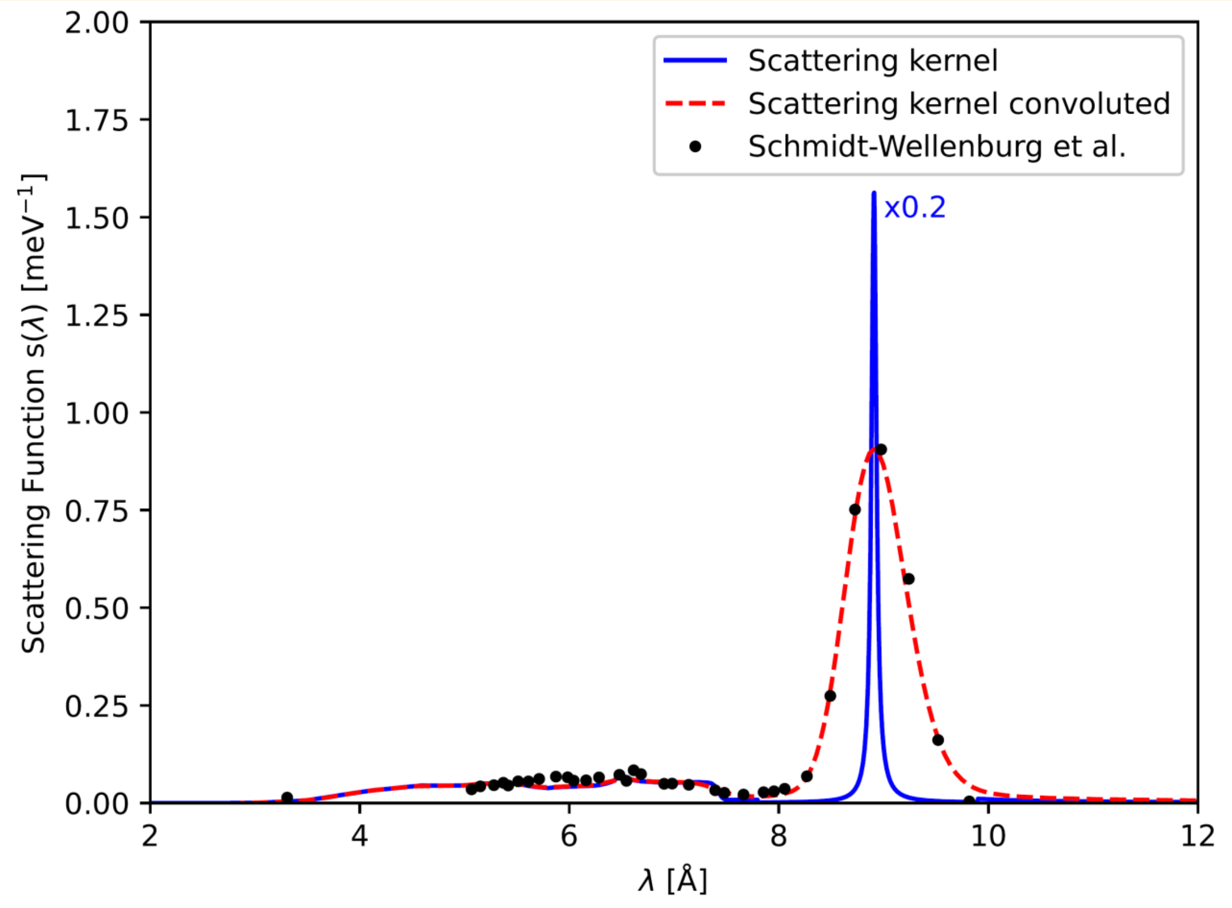



Data from K. Andersen Thesis (priv. Comm.)

# TOTAL SCATTERING CROSS SECTION



# ULTRA COLD NEUTRONS PRODUCTION




$$s(\lambda) = \hbar \int S(q, \hbar\omega)\delta(\hbar\omega - \hbar^2 k^2/2m_\mathrm{n})\,\mathrm{d}\omega$$

The derived function s(λ) at 1.24K compared to the measurements.
The convoluted curve was calculated using the resolution function.

# CONCLUSIONS

- A new scattering kernel to describe the interaction of slow neutrons with solid Deuterium has been developed.
- The main characteristics of this molecular solid are contained in the formalism, including dynamical aspects related to:
  - the lattice's density of states,
  - the Young-Koppel quantum treatment of the rotational motion,
  - the exact treatment of the one-phonon IA in the inelastic term,
  - the internal molecular vibration.
- The elastic processes involving coherent and incoherent contributions are also fully described, as well as the spin-correlation effects caused by the coupling of intrinsic and rotational angular momenta.

- We have developed a new model for the description of superfluid $^4He$ at low temperatures and programmed it into a custom version of NJOY2016 to produce thermal-scattering data.
- The model includes an exact description of the phonon-roton dispersion curve and a multi-phonon component, and we have shown that it reproduces well available measured cross-section and UCN production data.
- The current model can be used to create input that can be used together with NCrystal, either stand-alone or coupled together with a Monte-Carlo code, for calculations of UCN production from $^4He$ sources at low temperatures.

THANKS FOR YOUR ATTENTION!

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