DINS to probe nuclear quantum effects

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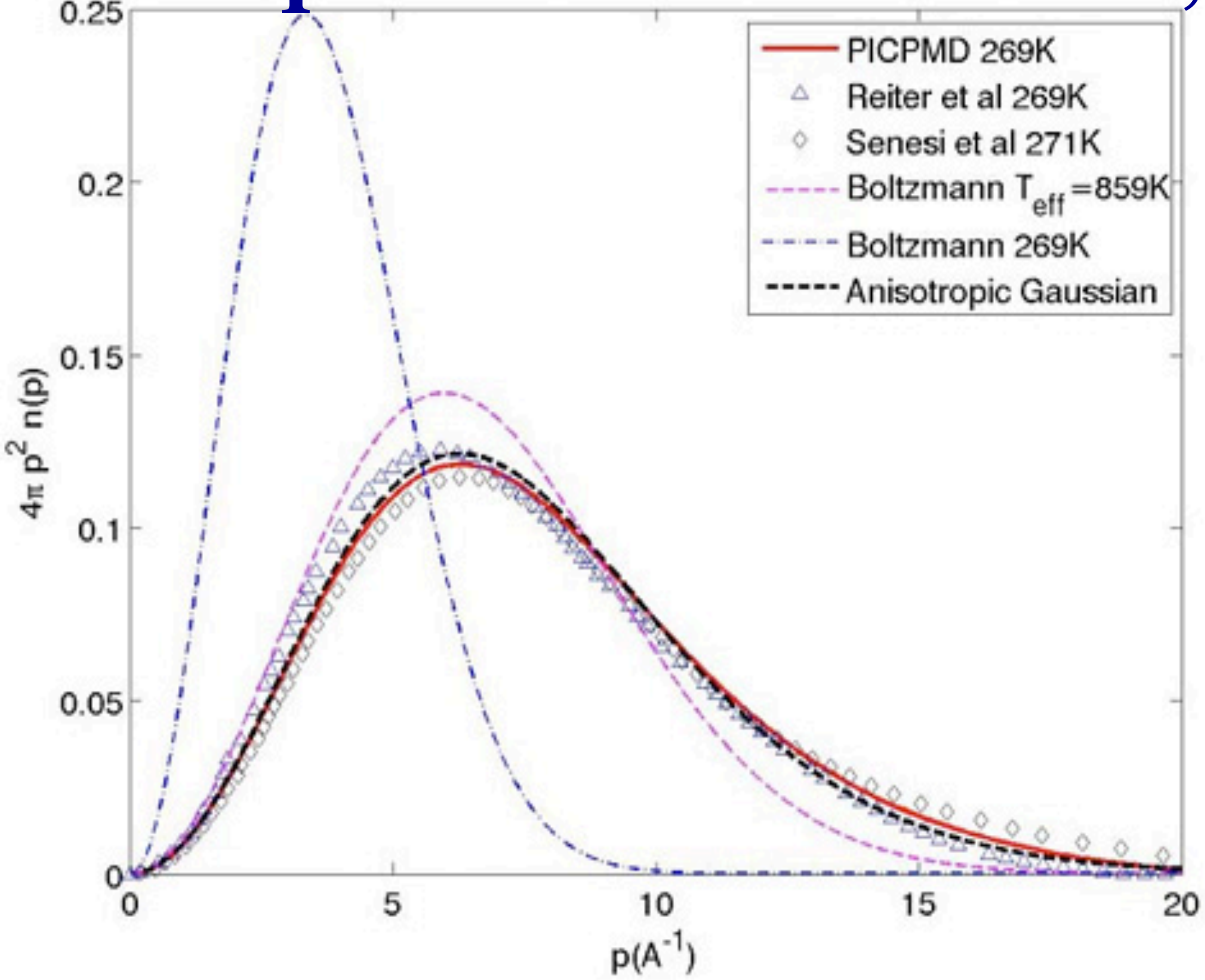
Water and Water Systems, Erice EMCSC 22 -31 July (2016)

Outline

- ❑ NQEs
- ❑ DINS (Compton Neutron Scattering)
  - ❑ Technique in brief (1985-2016)
  - ❑ Results
- ❑ Conclusions

Nuclear quantum effects in Ice, T=296K



The spherically averaged momentum distribution of the protons (shown here for ice at T=296 K) is quite different from the classical Maxwell-Boltzmann distribution at the same temperature. The path integral ab-initio molecular dynamics (PICPMD) result agrees well with two experiments (Reiter and Senesi). The Maxwell-Boltzmann distribution that fits better the PICPMD data has T=859 K. An even better fit is obtained with an anisotropic (multivariate) Gaussian distribution.

Courtesy of Roberto Car (Princeton University)

Nuclear Quantum Effects

$$\Delta x \, \Delta p \geq \hbar / 2$$

$$\frac{\langle p^2 \rangle}{2M} >> \frac{3}{2} k_B T$$

Nuclear Quantum Effects

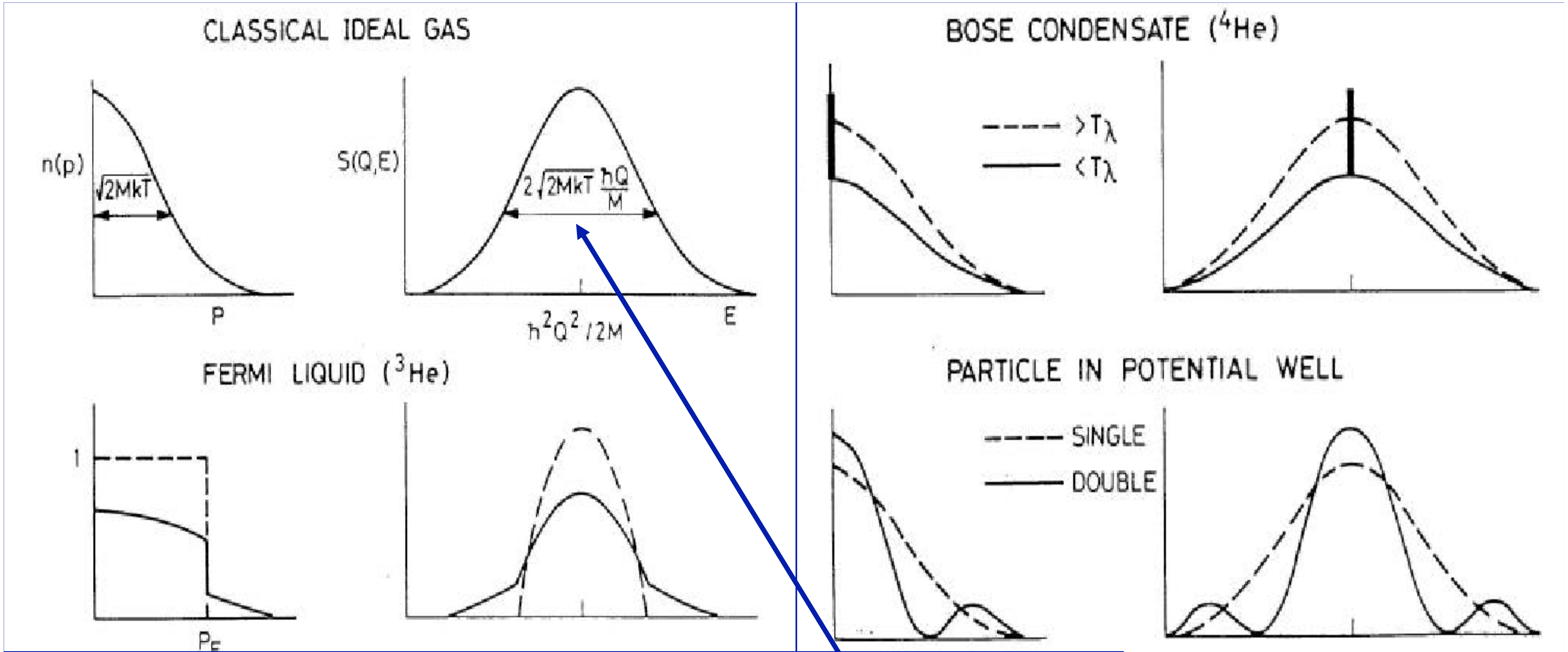
$\langle E_K \rangle$ & $n(p)$ → PES

$$n(\vec{p}) = \left| \int \psi(\vec{r}) \exp(i\vec{p} \cdot \vec{r}) d\vec{r} \right|^2$$

$$< E_k > = \frac{1}{2M} \int n(\mathbf{p}) p^2 d\mathbf{p}$$

{ **localization** } → **excess of $\langle E_K \rangle$**

$$\frac{\langle p^2 \rangle}{2M} >> \frac{3}{2} k_B T$$



$$n(\mathrm{p}) \propto \exp\left[-\mathrm{p}^2/(4Mk_BT)\right] \Rightarrow \sigma^2 = 2\mathrm{Mk_BT}$$

Classical system

Peak width of S(Q,ω) provides a direct measure of $\langle E_K \rangle$

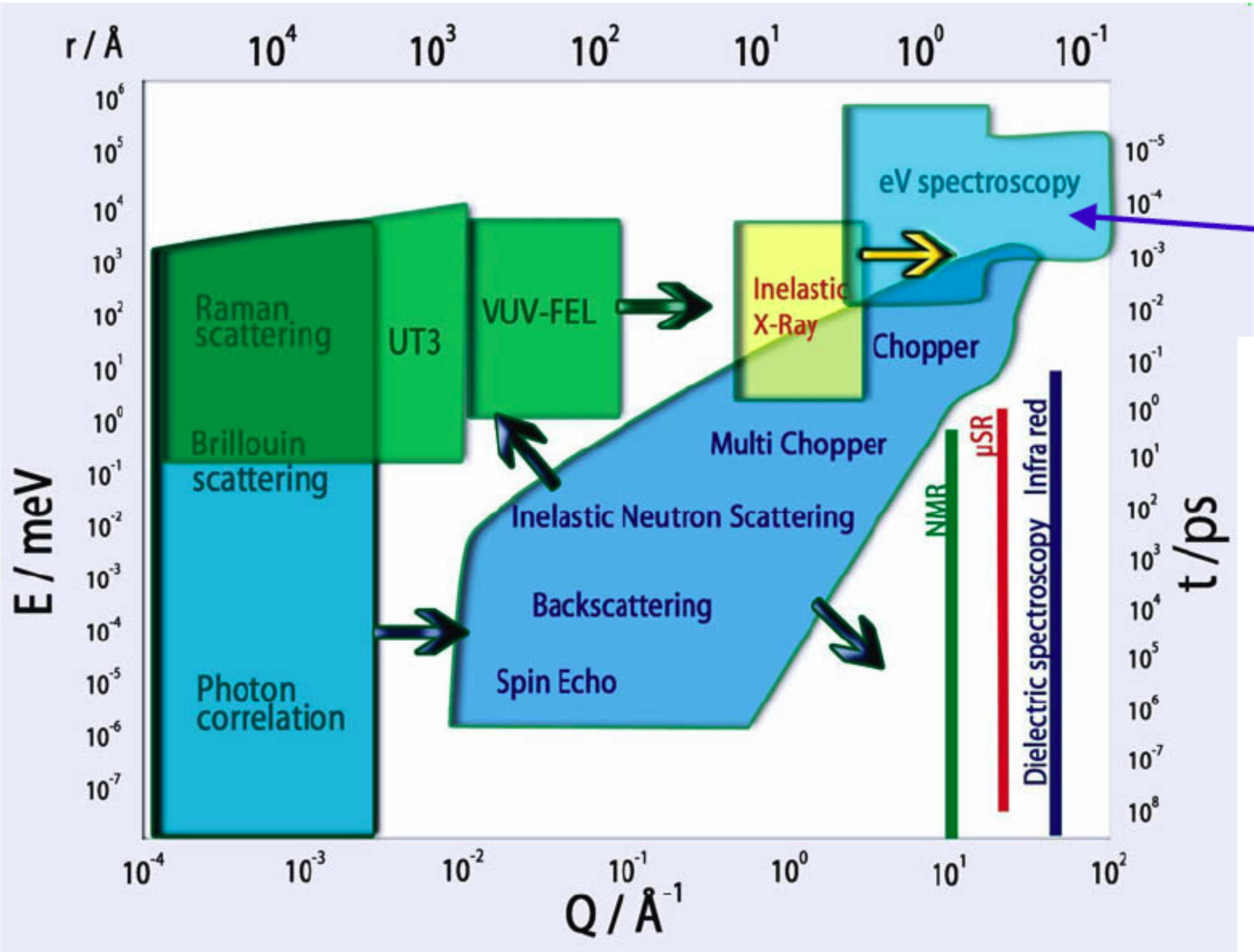
$$< E_k > = \frac{1}{2M}\int n(\boldsymbol{p})\,p^2\,d\boldsymbol{p}$$

$$n(\vec{p}) = \left|\int \psi(\vec{r})\exp(i\vec{p}\cdot\vec{r})\,d\vec{r}\right|^2$$

Momentum Distribution is "Diffraction Pattern" of Wave function

J. M. F. Gunn, C. Andreani, J. Mayers, *J. P. C: Solid State Physics* **19**, L835 (1986)

Struck Atoms travel only a short time- small distance



implying large Q, ω

$20\ Å^{-1} < q < 250\ Å^{-1}$

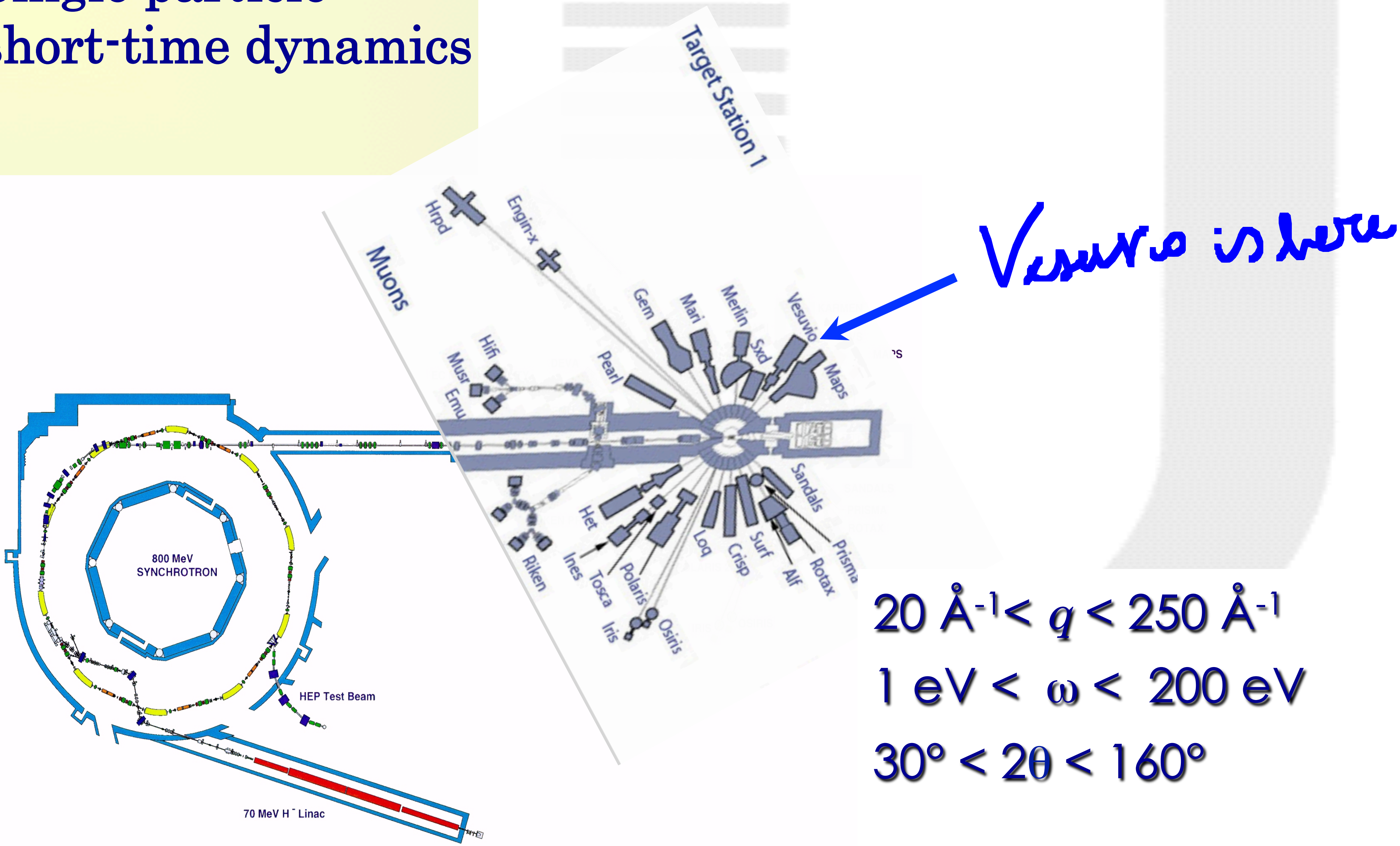
$\omega > 1\,eV$

$10^{-5}\,ps < t < 10^{-3}\,ps$

$0.1\ Å < r < 0.2\ Å$

VESUVIO at ISIS Neutron Source (UK)

Single particle
short-time dynamics



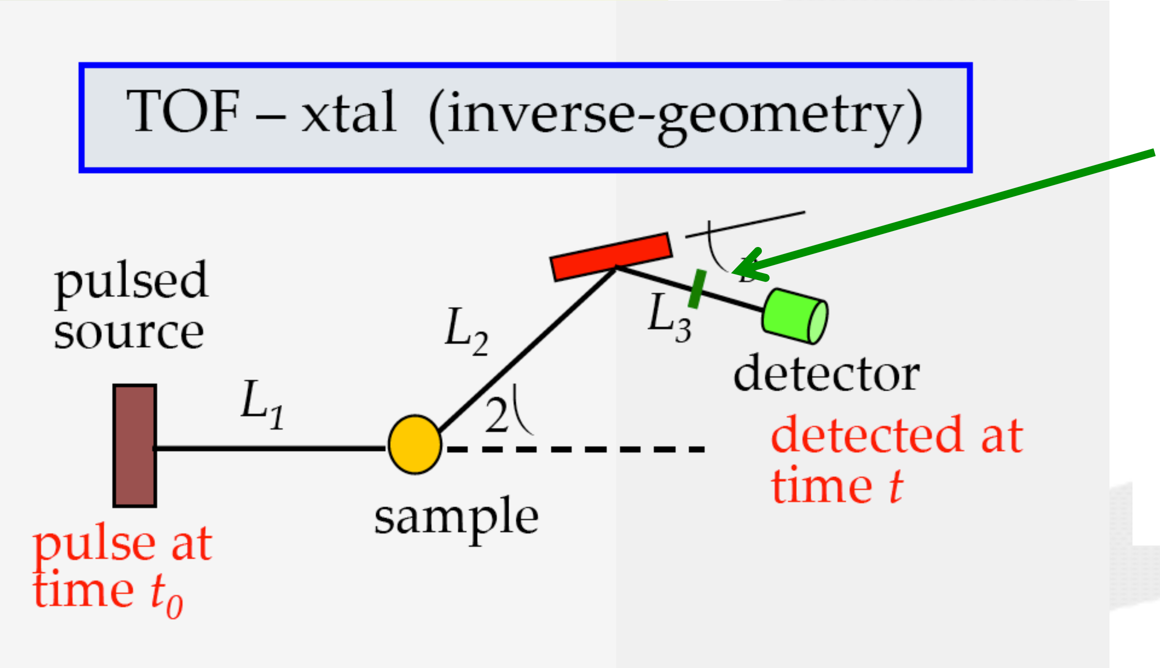
$20\ \text{Å}^{-1} < q < 250\ \text{Å}^{-1}$

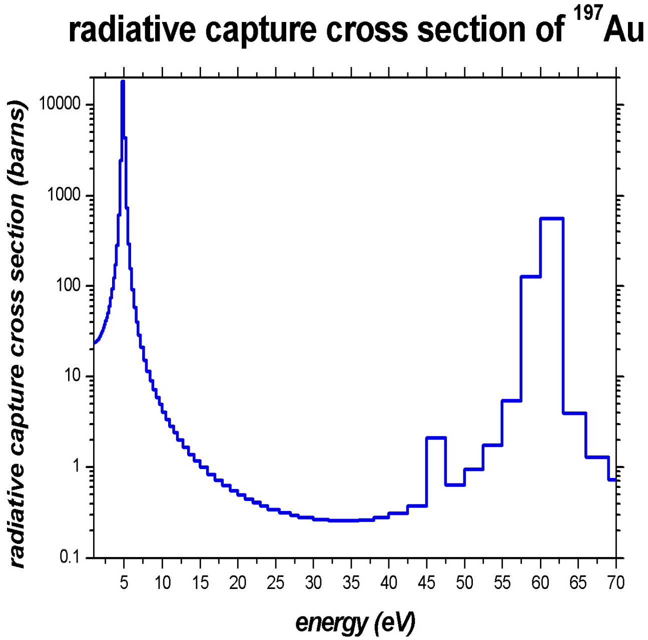
$1\ \text{eV} < \omega < 200\ \text{eV}$

$30° < 2\theta < 160°$

VESUVIO is a filter spectrometer (at eV energy)

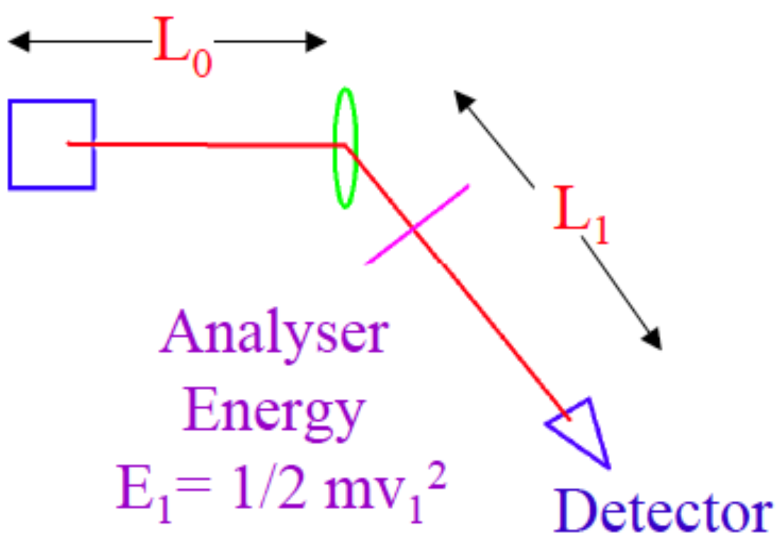
- Indirect geometry spectrometer
- Scattered neutron energy is selected by filters
- Incident neutron energy is determined by time-of-flight





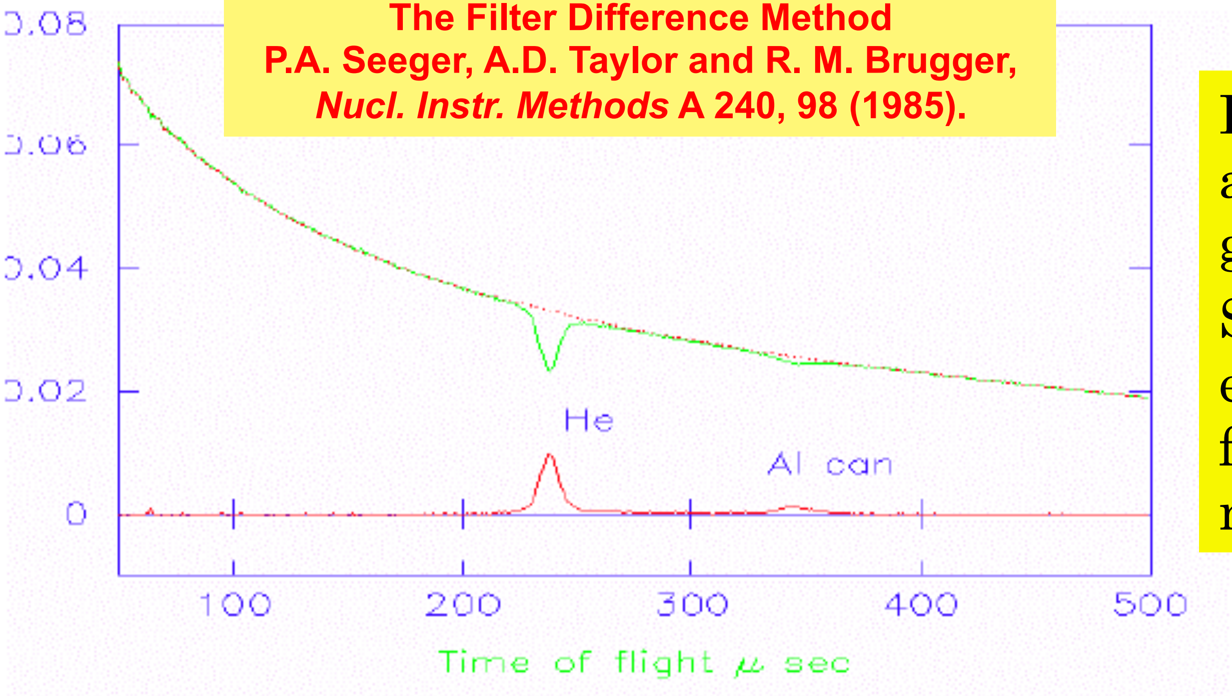
RFS

$$t = \frac{L_1}{v_1} + \frac{L_0}{v_0}$$



$$\hbar\omega = \tfrac{1}{2} m\left(v_0^{\ 2} - v_1^{\ 2}\right)$$

$$q^2 = m^2\left(v_0^{\ 2} + v_1^{\ 2} - 2 v_0 v_1 \cos\theta\right)$$



The Filter Difference Method
P.A. Seeger, A.D. Taylor and R. M. Brugger, *Nucl. Instr. Methods* A 240, 98 (1985).

Incident neutrons of all available energies are generated at t=0

Scattered neutrons are energy selected by the filters and recorded by neutron detector

DINS

- **Inelastic neutron scattering cross section expressed in terms of n(p) → Impulse Approximation**

$$\frac{\hbar q}{m} S_{\mathrm{IA}}(\mathbf{q},\omega) = J_{\mathrm{IA}}(y,\hat{\mathbf{q}}) = \int n(\mathbf{p})\delta\left(y - \mathbf{p}\cdot\hat{\mathbf{q}}\right)\mathrm{d}\mathbf{p}$$

↑

Responce Function or
Neutron Compton Profile

$$y = \frac{m}{\hbar q}\left[\omega - \frac{\hbar q^2}{2m}\right]$$

J. M. F. Gunn, C. Andreani, J. Mayers, *J. P. C: Solid State Physics* **19**, L835 (1986)

Neutron Compton Profile

$$J_{\mathrm{IA}}(y) = 2\pi \int_{|y|}^{\infty} p n(p) \mathrm{d}p$$

NCP broadened by:

- **Terms at finite-q, due to departure from IA;**
- **Experimental resolution function**

$$F(y,q) = [J_{\mathrm{IA}}(y) + \Delta J(y,q)] \star R(y,q)$$

VESUVIO (eVS upgrade)

The Resonance Filter Spectrometer (1982-2003)

- **Filter Difference technique (D)**
- **Double Difference Filter technique (DD)**

Resonance Detector Spectrometer (2002)

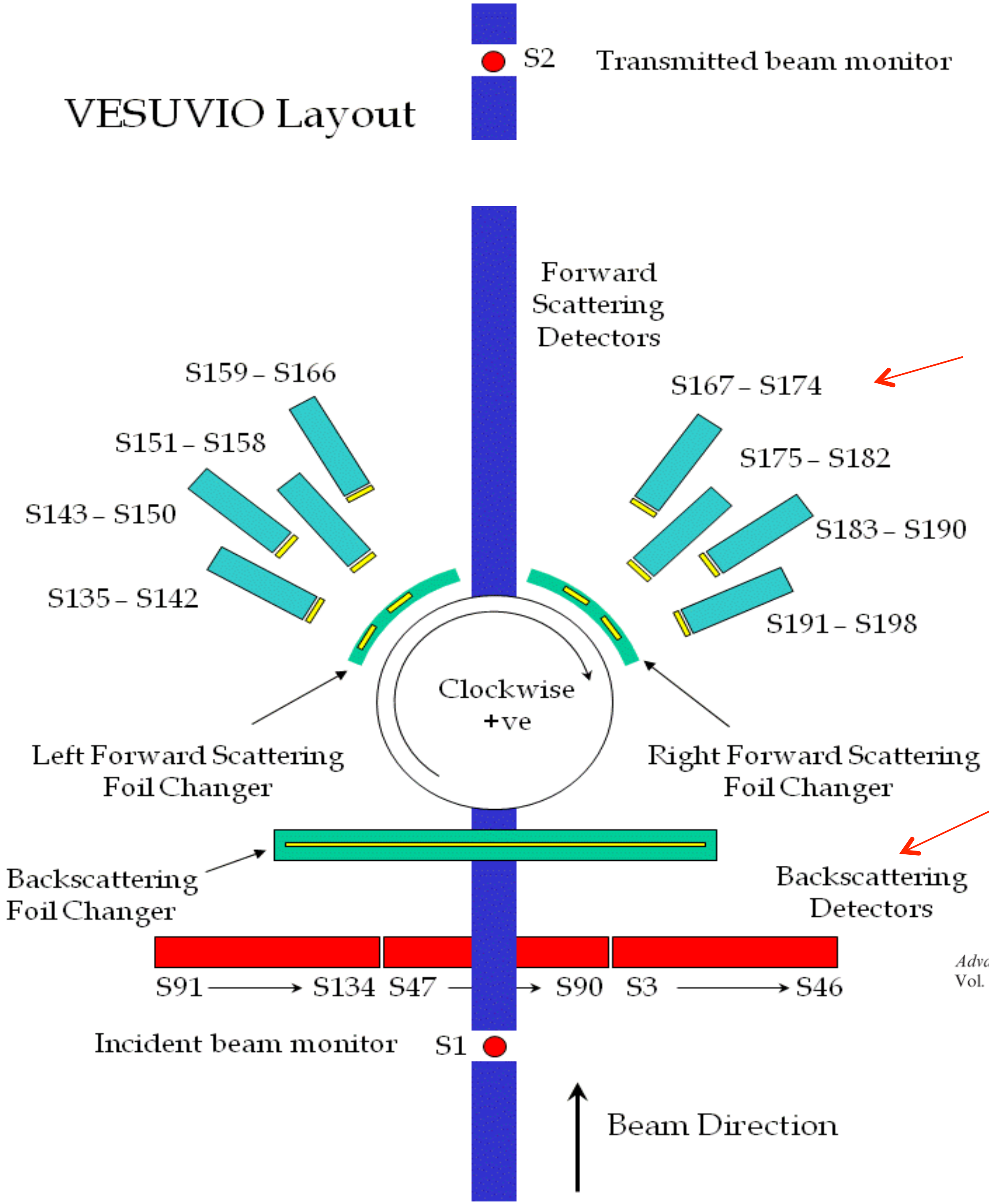
- Foil cycling technique (FC)

RDS & RFS

YAP detector + ^{6}Li neutron detectors (2006)

- Due to installation of YAP detectors VESUVIO has gained one order of magnitude better accuracy for proton measurements
- Accuracy in widths of $n_H(p)$ is ~ 0.5%

VESUVIO from 2006 operates in RDS & RFS



YAP gamma detectors

^{6}Li neutron detectors

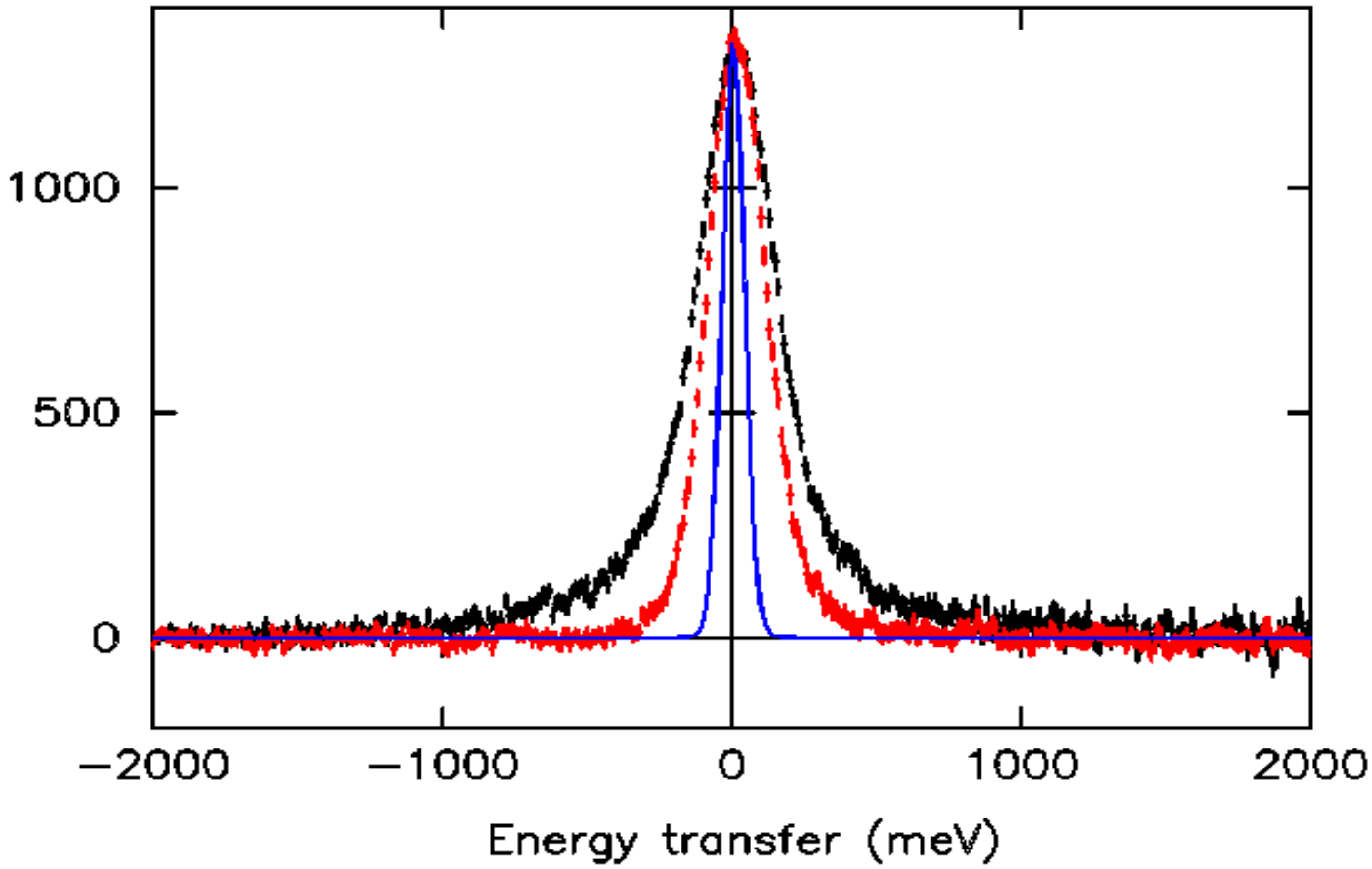
Advances in Physics,
Vol. 54, No. 5, July–August 2005, 377–469

Taylor & Francis
Taylor & Francis Group

Measurement of momentum distribution of light atoms and molecules in condensed matter systems using inelastic neutron scattering

C. ANDREANI*†, D. COLOGNESI‡, J. MAYERS§, G. F. REITER¶ and R. SENESI†||

Resolution improvement FC versus FD

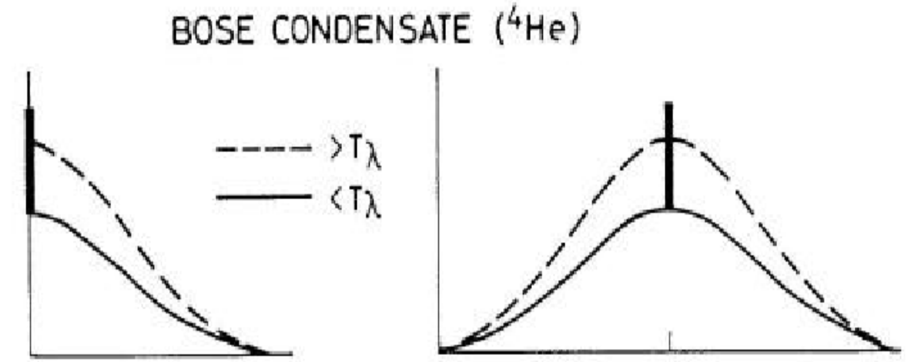


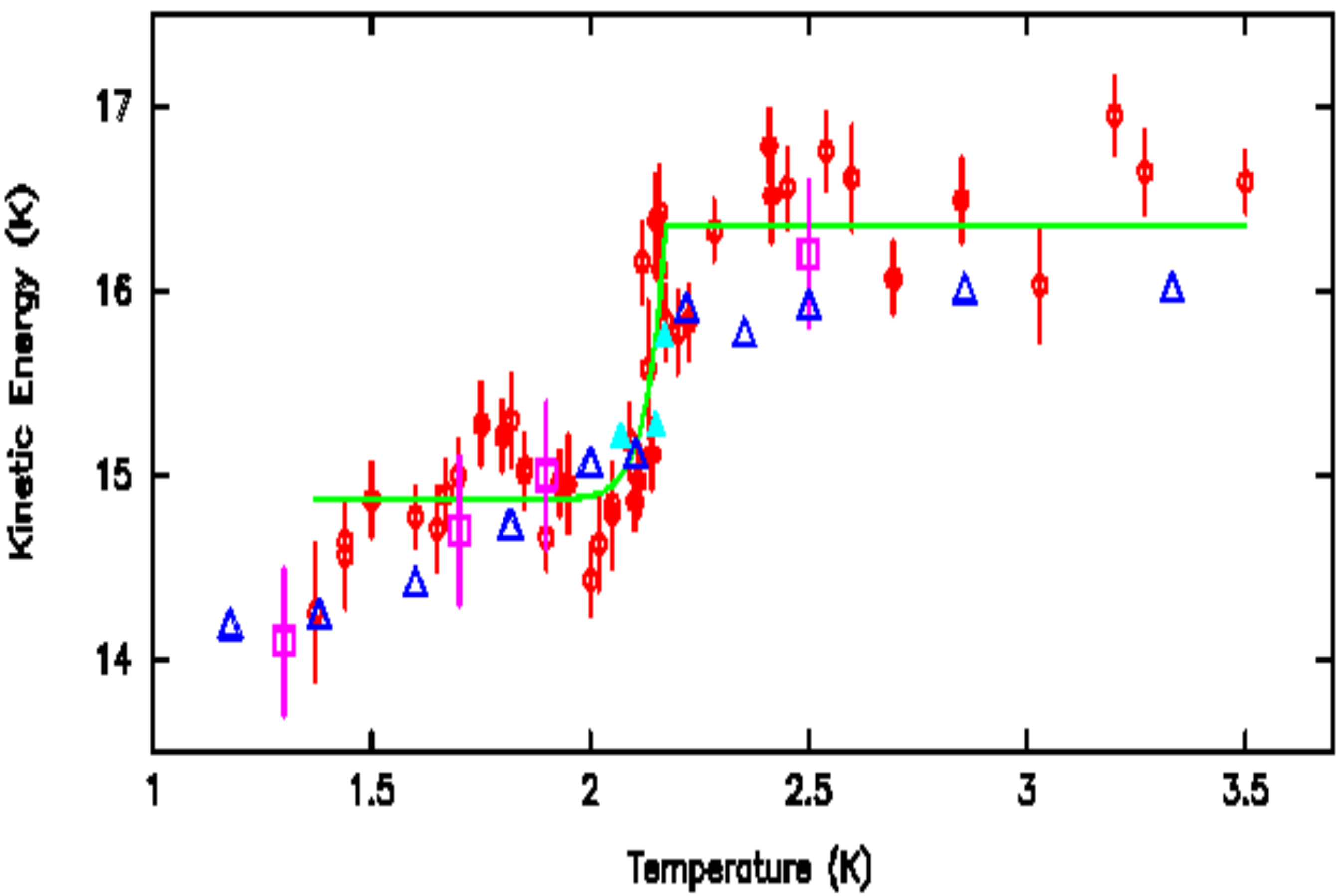
YAP same resolution as Li-glass in DD (< 2006)

A selection of DINS Measurements

Mean Kinetic Energy of ^{4}He

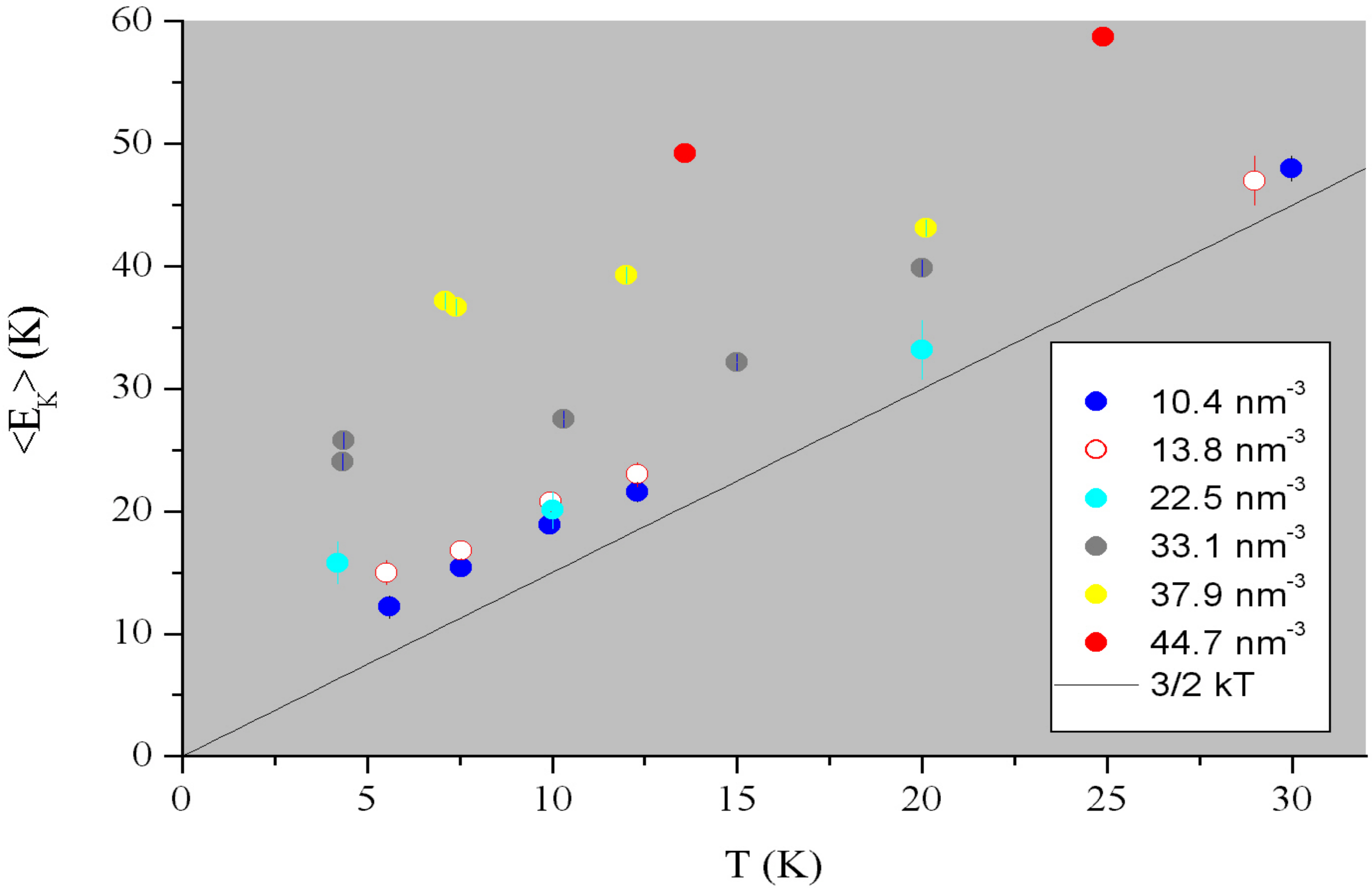
DINS (and INS)

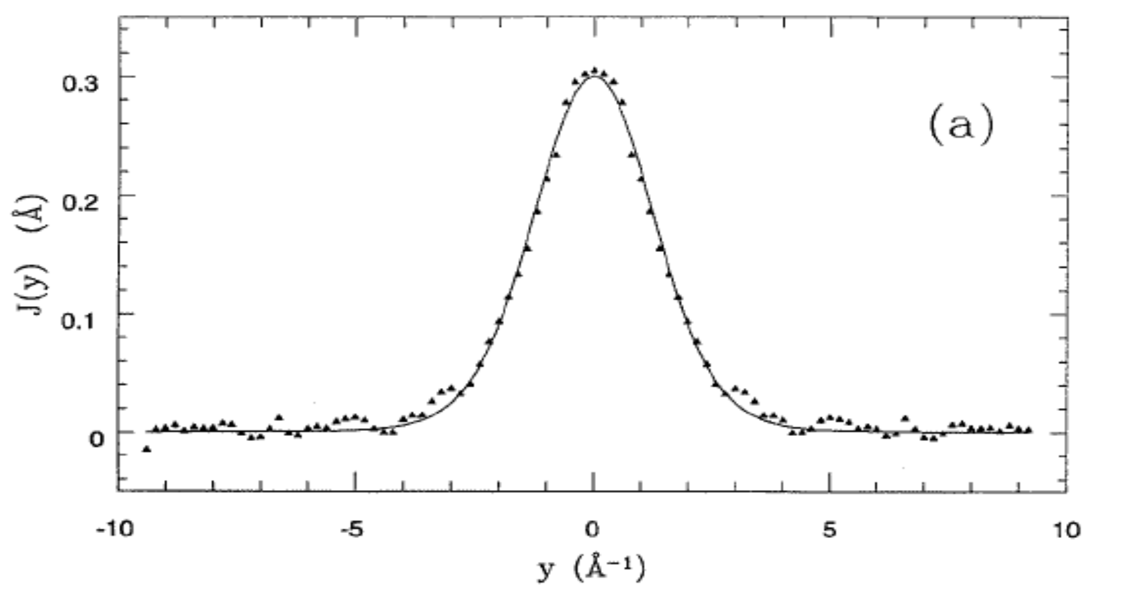




We

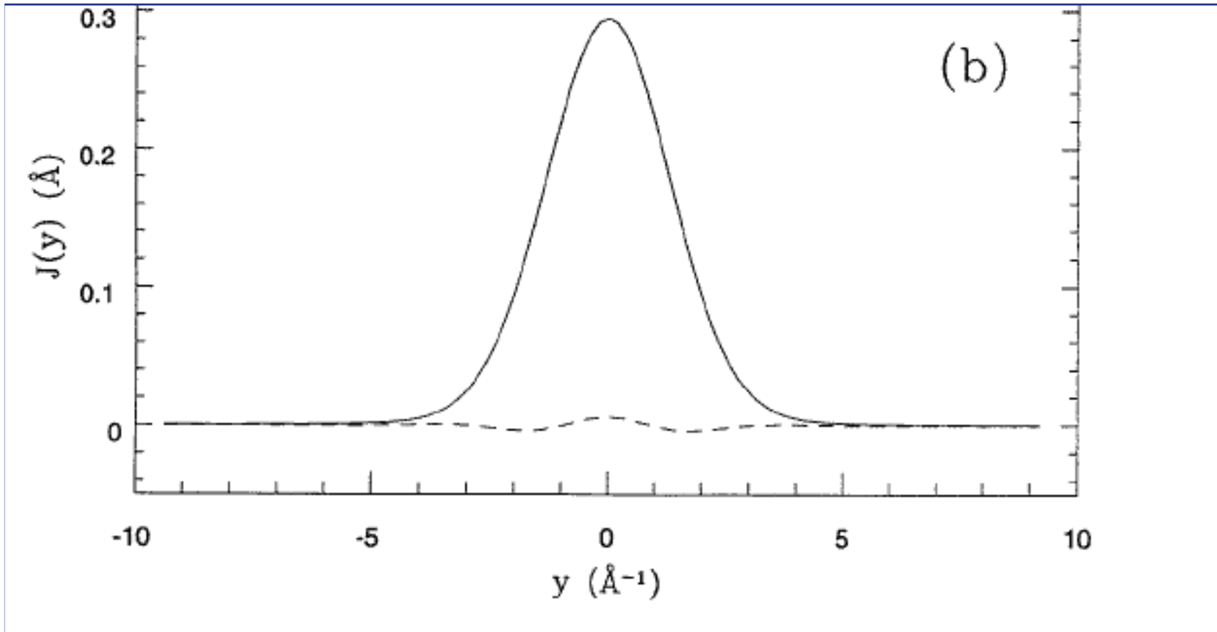
Liquid ^{4}He $\langle E_K \rangle$ > 3/2 KT!





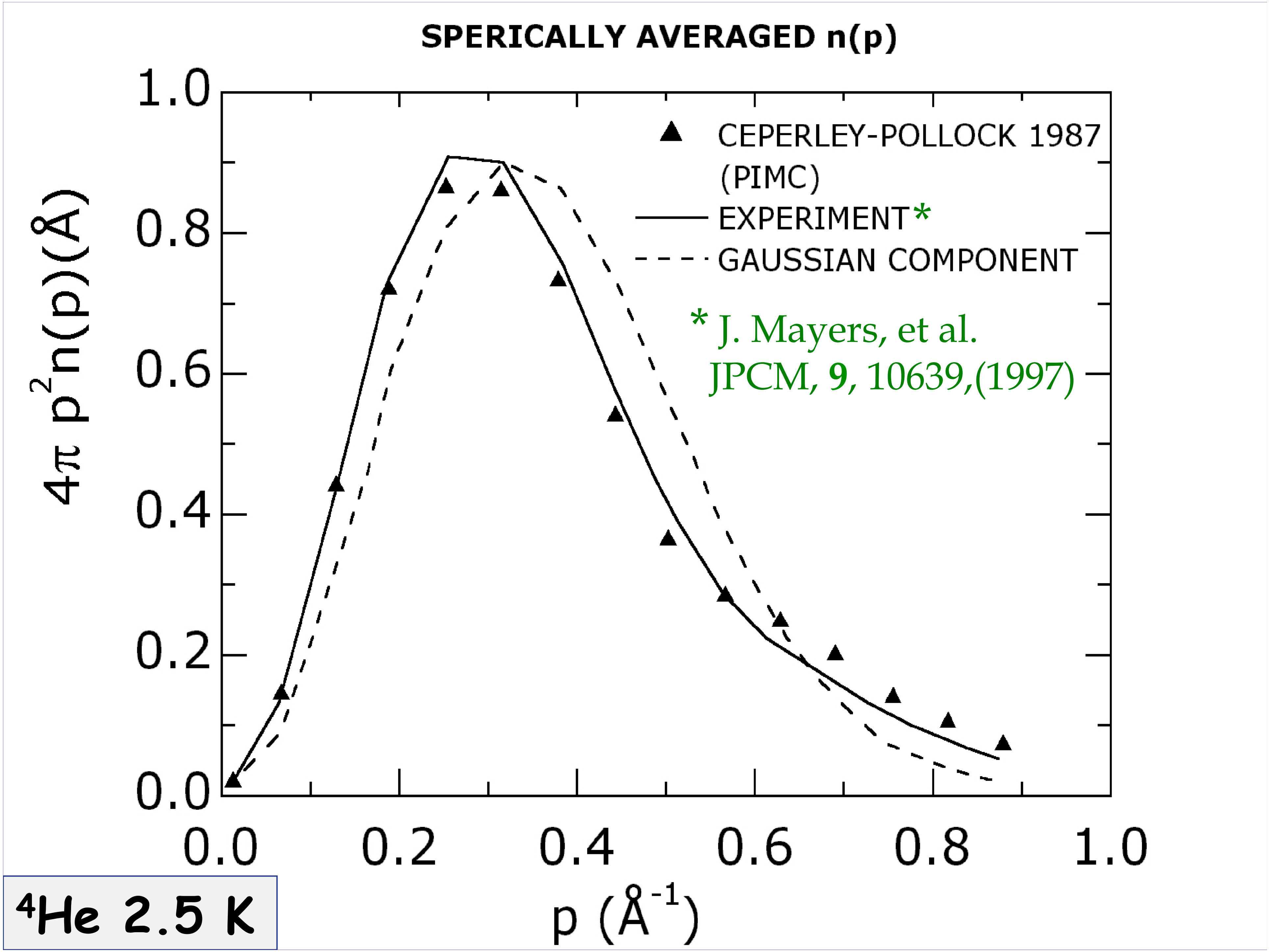
^{4}He at 2.5 K

Figure 3. (a) The response function $J(y)$ for ^{4}He at 2.5 K from the cooled U filter: triangles are the experimental data; the solid line is the result of the fit. (b) The solid line is the Gaussian component of the fit; the dashed line is the non-Gaussian component, with both including a resolution contribution.

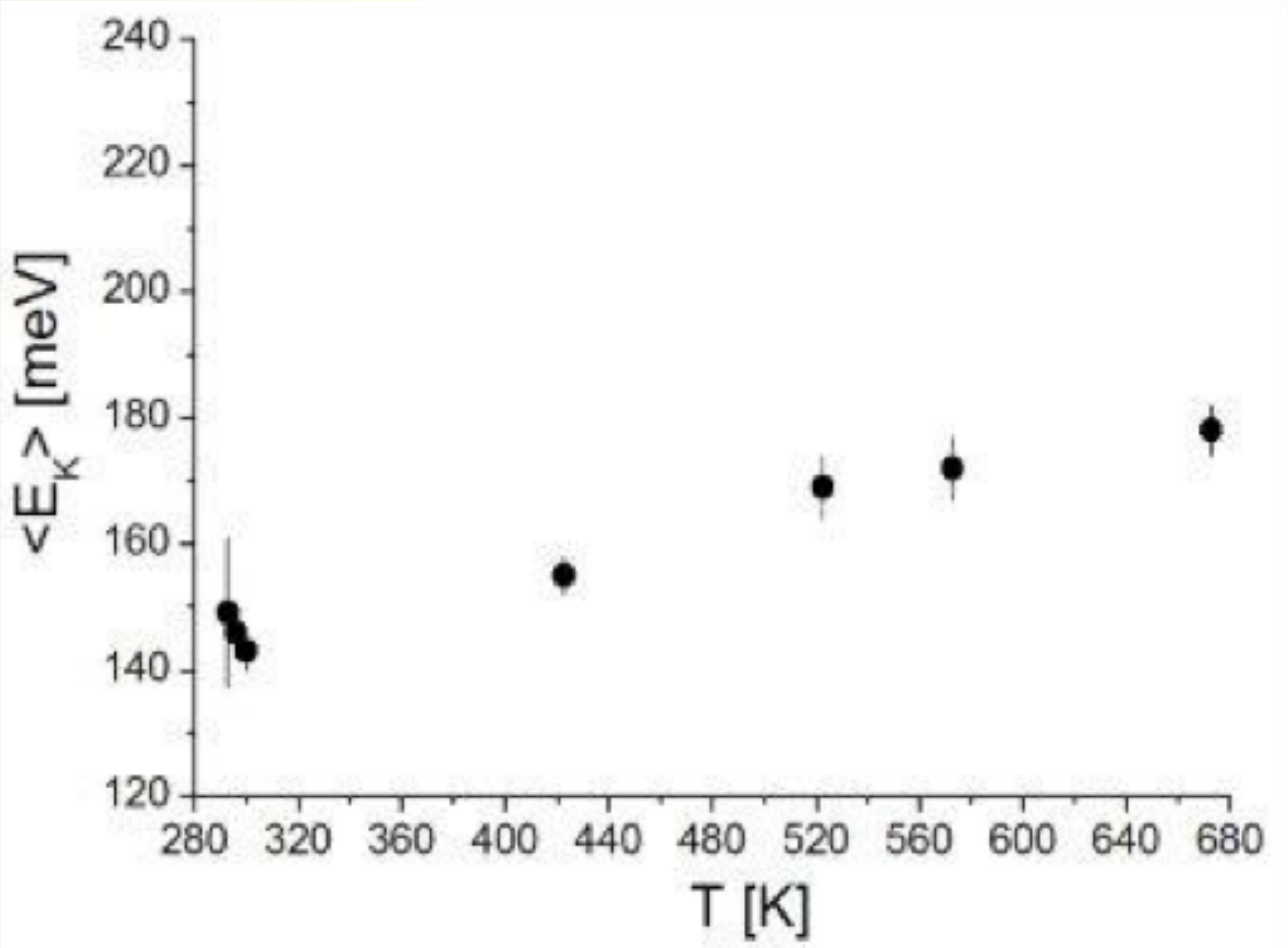


$\delta = 0.63 \pm 0.06$

* J. Mayers, et al.
JPCM, **9**, 10639,(1997)

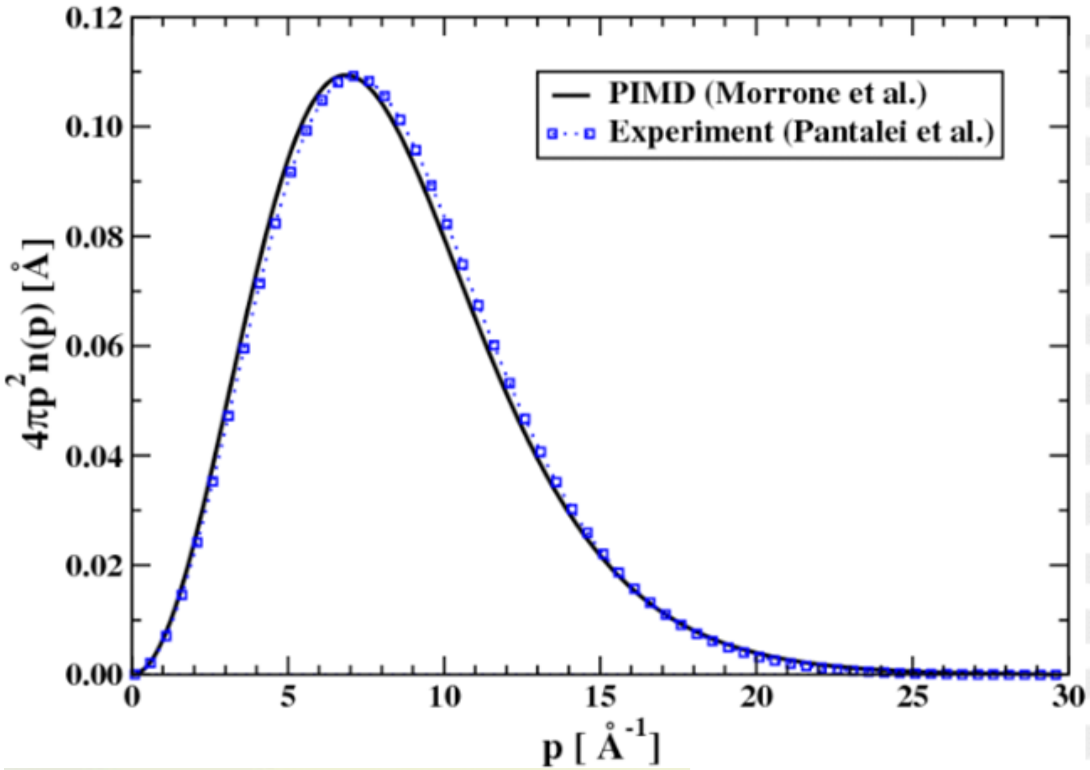

SPERICALLY AVERAGED n(p)
CEPERLEY-POLLOCK 1987 (PIMC)
EXPERIMENT*
GAUSSIAN COMPONENT
* J. Mayers, et al. JPCM, 9, 10639,(1997)
4π p²n(p)(Å)
p (Å⁻¹)
1.0
0.8
0.6
0.4
0.2
0.0
0.0
0.2
0.4
0.6
0.8
1.0
⁴He 2.5 K

Supercritical water
n(p) very similar to the H_2O monomer!



C. Pantalei et al. Phys Rev Letters **100** 177801 (2008)

Supercritical water
n(p) very similar to the H_2O monomer!



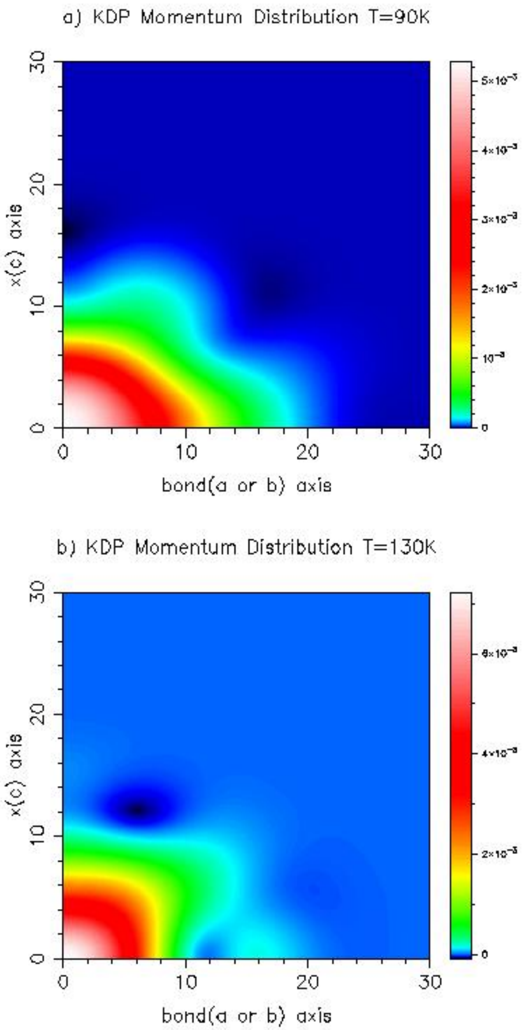
DINS Exp: C. Pantalei et al. PRL **100** 177801 (2008):
T= 673 K
P=106 MPa
r=0.7 g/cm^3

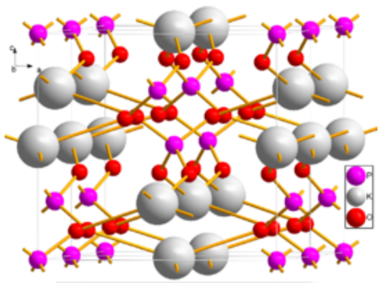
Theory: J. Morrone et al, JCP **126**, 234504 (2007)

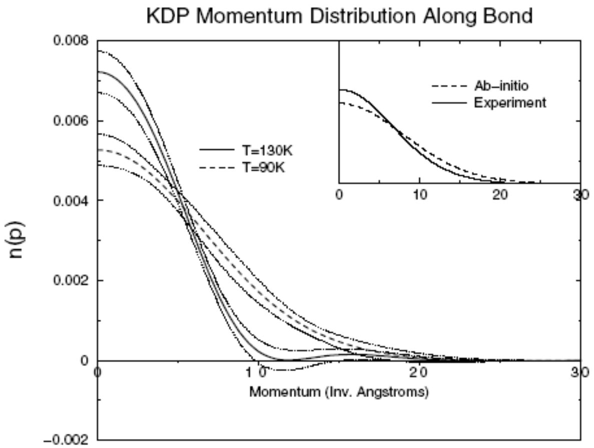
n(p) harmonic- anisotropic lineshape :

$$n(p) = \left\langle \Pi_i \frac{1}{\sqrt{2\pi\,\sigma_i^{\,2}}} \exp\left(- \frac{p^2}{2\,\sigma_i^{\,2}} \right) \right\rangle_\Omega$$

High-energy Neutrons as the Ultimate *Wavefunction Diffractometer*

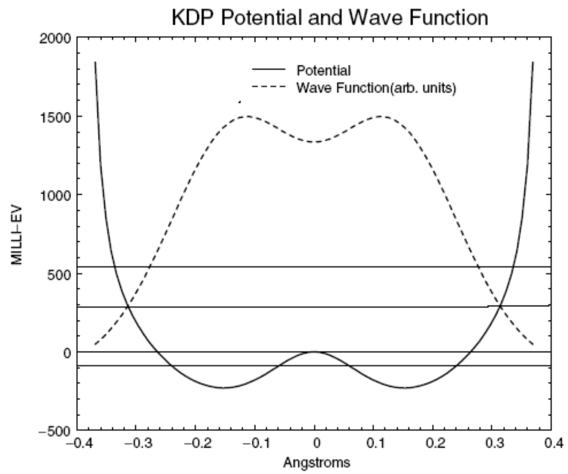






$$n(p) \sim \left| \int \Psi(\vec{\mathbf{r}}) e^{i\vec{\mathbf{p}}\cdot\vec{\mathbf{r}}} \, d\vec{\mathbf{r}} \right|^2$$

$$V(\vec{\mathbf{r}}) = E - \frac{\int \left(\mathbf{p}^2 / 2m \right) \sqrt{n(\vec{\mathbf{p}})} e^{i\vec{\mathbf{p}}\cdot\vec{\mathbf{r}}} \, d\vec{\mathbf{p}}}{\int \sqrt{n(\vec{\mathbf{p}})} e^{i\vec{\mathbf{p}}\cdot\vec{\mathbf{r}}} \, d\vec{\mathbf{p}}}$$



Requires single crystals

*G. Reiter Phys Rev Lett **89** 135505 (2002)*

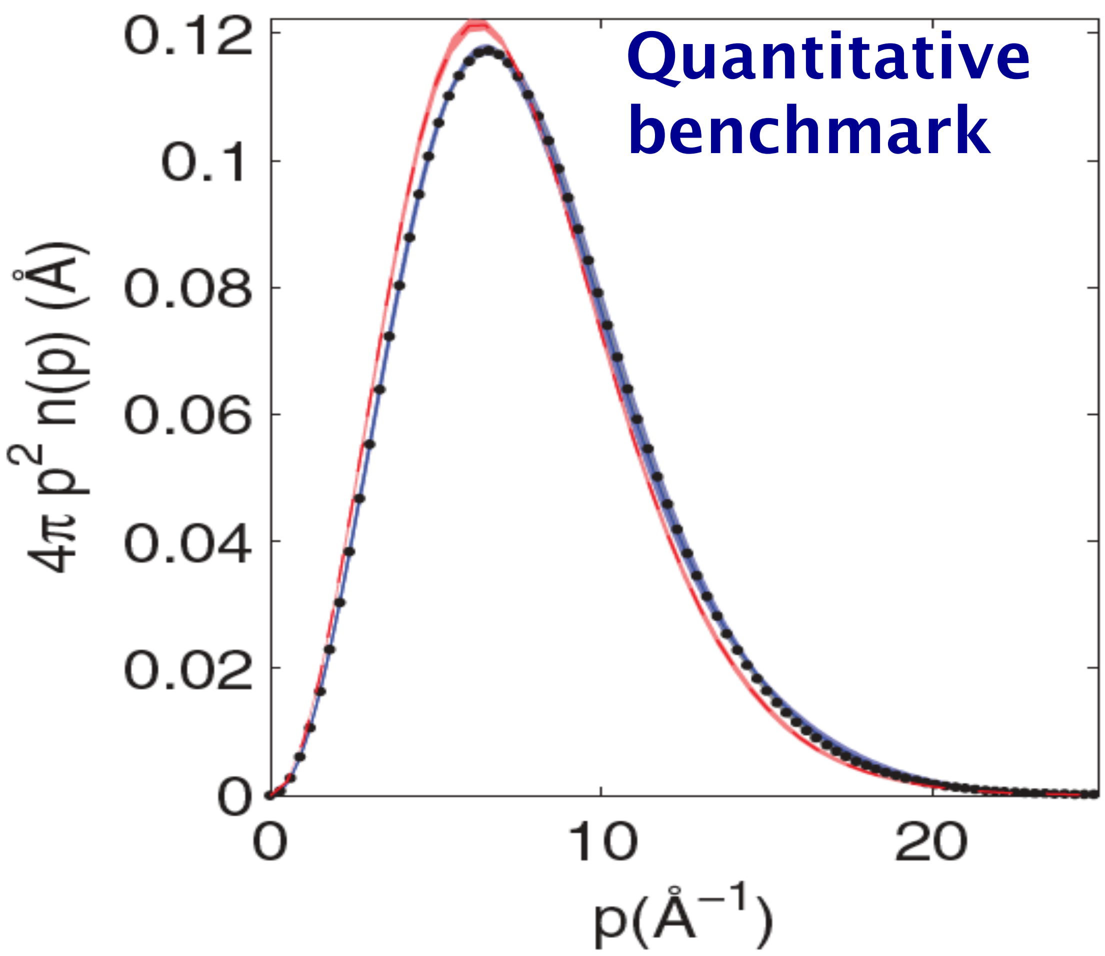
ICE

DINS ● at 271 K

$<E_K> = 154 \pm 2$ meV

PICPMD --- at 269 K

$<E_K> = 143 \pm 2$ meV



** D. Flammini, A. Petropaolo, R. Senesi, C. Andreani, F. McBride, A. Hodgson, M. Adams, L. Lin, R. Car, J. Chem. Phys.* 136, 024504 (2012)

n(p) and <E_K> in H_2O

- Spherical average anisotropic Gaussian distribution:

$$4\pi p^2 n(p) = \left\langle \frac{\delta(p - |\mathbf{p}|)}{\sqrt{8\pi^3}\sigma_x\sigma_y\sigma_z} \exp\left(-\frac{p_x^2}{2\sigma_x^2} - \frac{p_y^2}{2\sigma_y^2} - \frac{p_z^2}{2\sigma_z^2}\right)\right\rangle$$

- x,y and z → the three molecular axes of the H_2O molecules

n(p) and <E$_K$>

This expression involves three parameters σ_α^2 related

- to principal frequencies ω_α by:

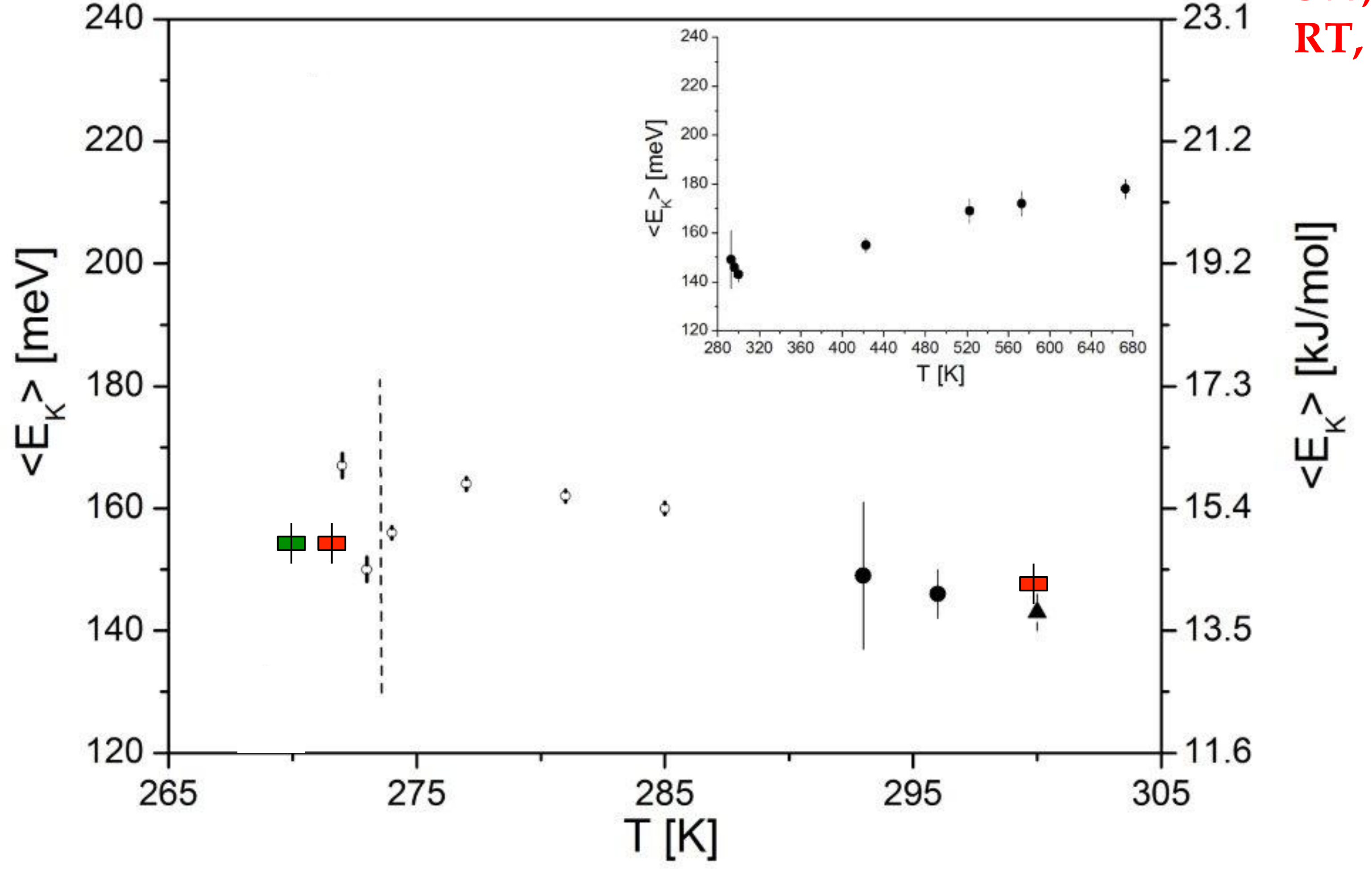
$$\sigma_\alpha^2 = \frac{m\omega_\alpha}{2\hbar}\coth\frac{\beta\hbar\omega_\alpha}{2} \qquad \alpha = x, y, z$$

- and:

$$\langle E_\alpha \rangle = \hbar^2\sigma_\alpha^2/2m$$

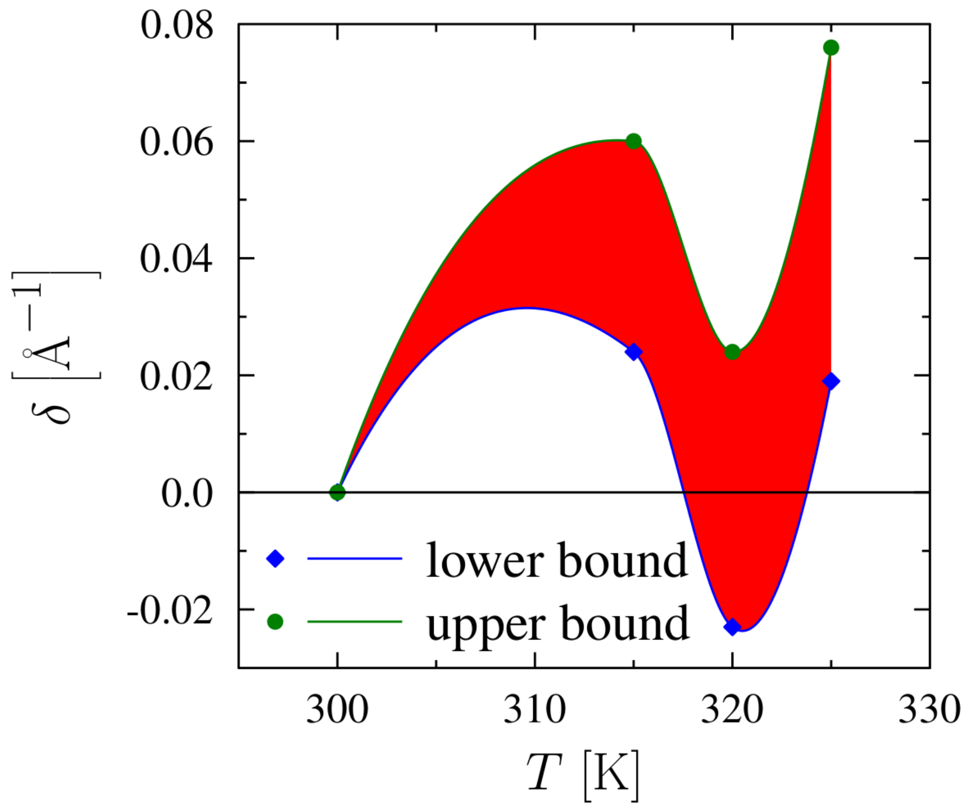
H_2O: $<E_K>$ *vs* T

ICE:
T=271 K
H_2O:
SW, T= 271 K
RT, T=300 K



C. Andreani, *et al*. JPC*L* **7** (12), 2216–2220 (2016); D. Flammini, JCP **136**, 024504 (2012)
C. Andreani, *et al*. JCP, **115**, 11243 (2001), C. Pantalei et al. PRL**, 100,** 177801 (2008)
A. Pietropaolo *et al*., PRL, **100**, 127802 (2009),
A. Pietropaolo *et al*., *Braz. J. Phys*, **39**, 321 (2009)

Proton quantum dynamics of water across 315 K
(F. Mallamace et al. 2016)



Temperatures

300 K

315 K

320 K

325 K

Acknowledgments

Roberto Car	Princeton University
Michele Ceriotti	Ecole Polytechnique Federale de Lausanne
Andrew Hodgson	University of Liverpool
Lin Lin	Lawrence Berkeley National Lab.
Francesco Mallamace	University of Calabria
Fiona McBride	University of Liverpool
George Reiter	University of Huston
Giovanni Romanelli	Rome Tor Vergata and ISIS Neutron Source
Roberto Senesi	Rome Tor Vergata

THANK YOU

QUESTIONS?