Filter Spectrometers

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Carla Andreani

What we will cover

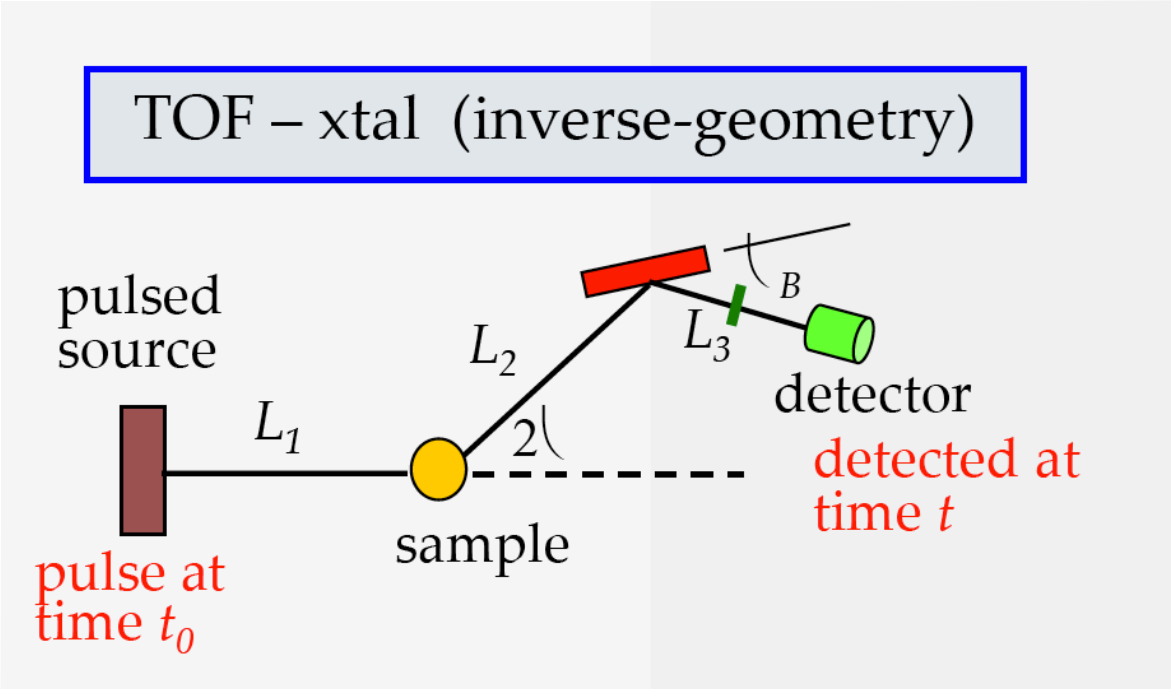
- **Filter Spectrometers – basics**
  - Kinematics
  - DINS
  - The Resonance Filter Spectrometer
    - Filter Difference Technique (FD)
    - Double Difference Technique (DDT)
  - Resolution Components
  - Resonance Detector Spectrometer
    - Cycling Technique (CT)
- **What do we measure and why**
- **Examples**

Filter Spectrometers – basics

…

Filter spectrometers at eV energy

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



Time of flight neutron measurements

Fixed v_1 (final v)
(Inverse Geometry)

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

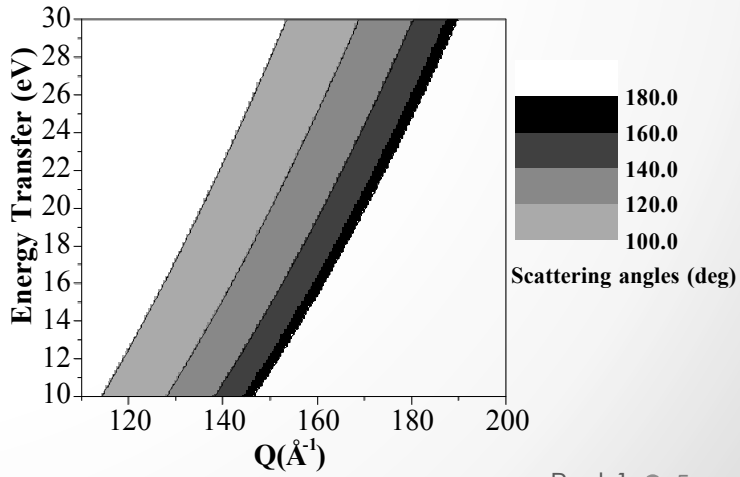
$$k_0 = \frac{mv_0}{\hbar} \qquad E_0 = \tfrac{1}{2}mv_0^2 \qquad k_1 = \frac{mv_1}{\hbar} \qquad E_1 = \tfrac{1}{2}mv_1^2$$

$$q^2 = k_1^2 + k_2^2 - 2k_1k_2\cos\theta$$

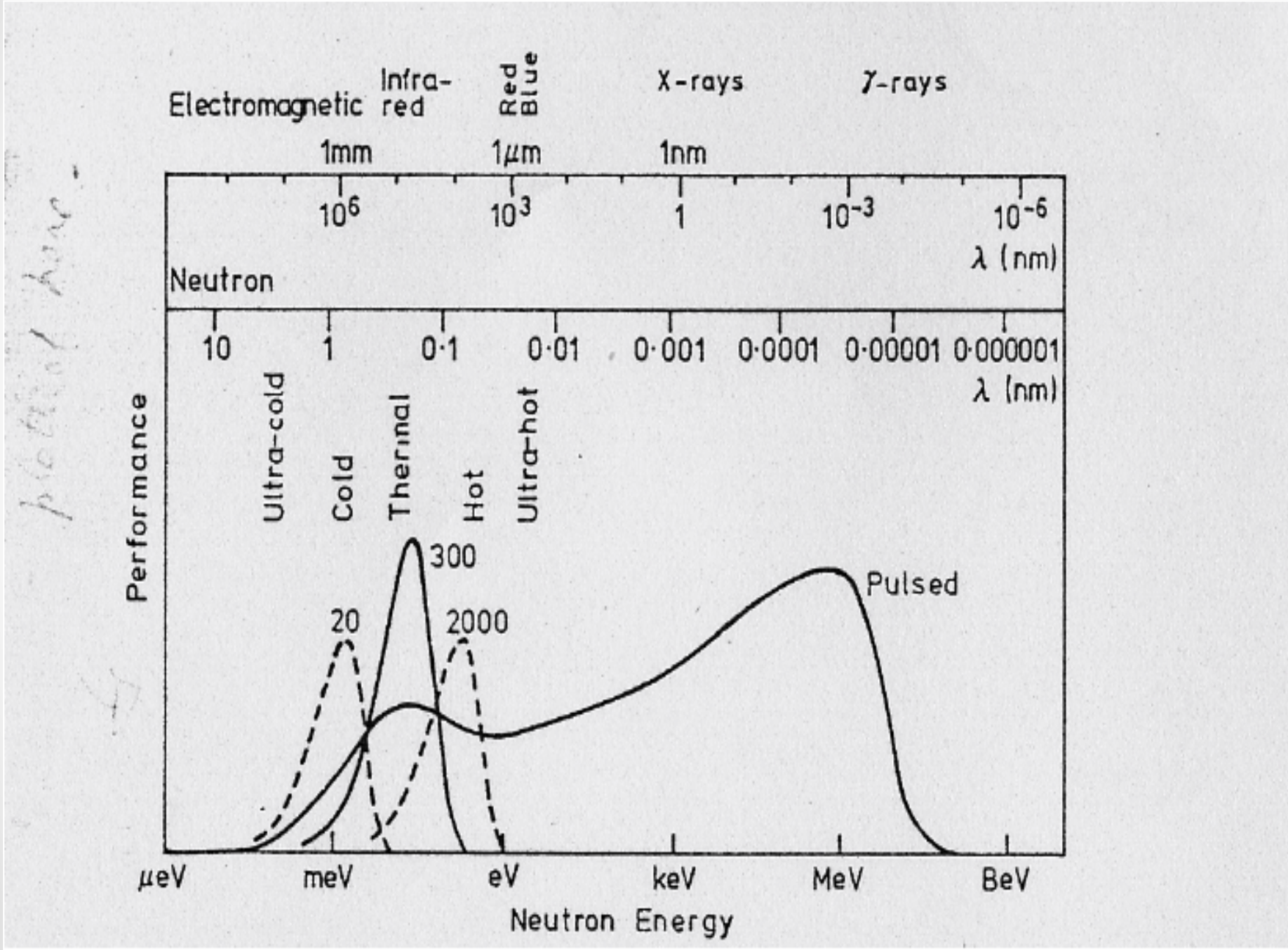
Wave vector transfer

$$\omega = E_0 - E_1$$

Energy transfer



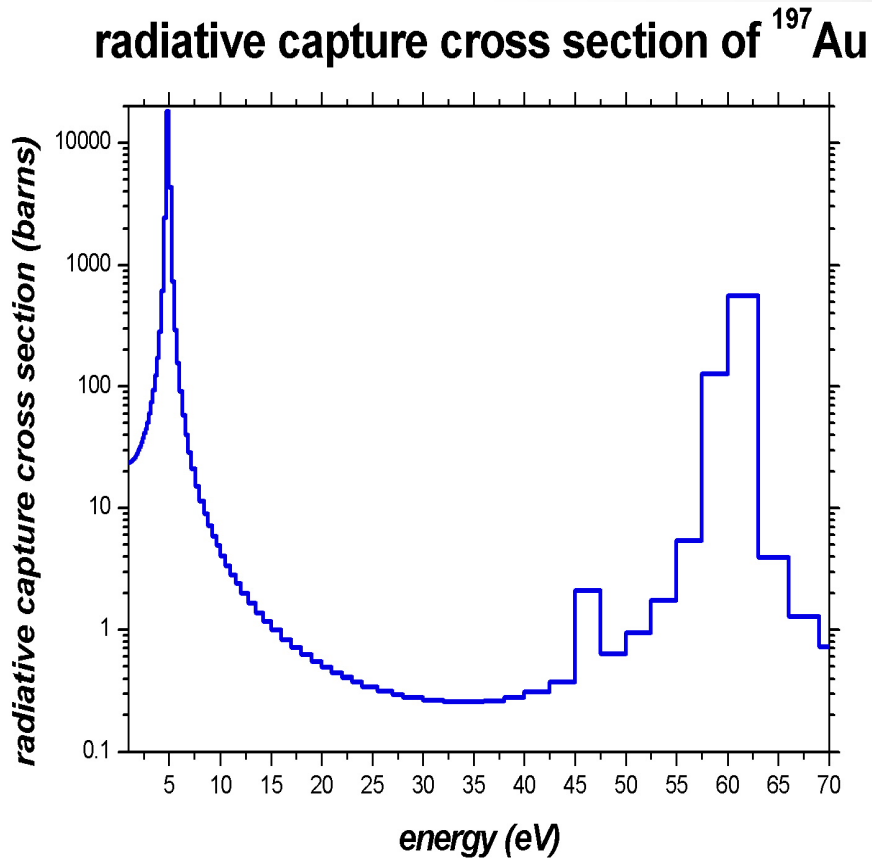
Time of flight measurements with eV neutrons



How to select the final neutron energy in the 1-150 eV range

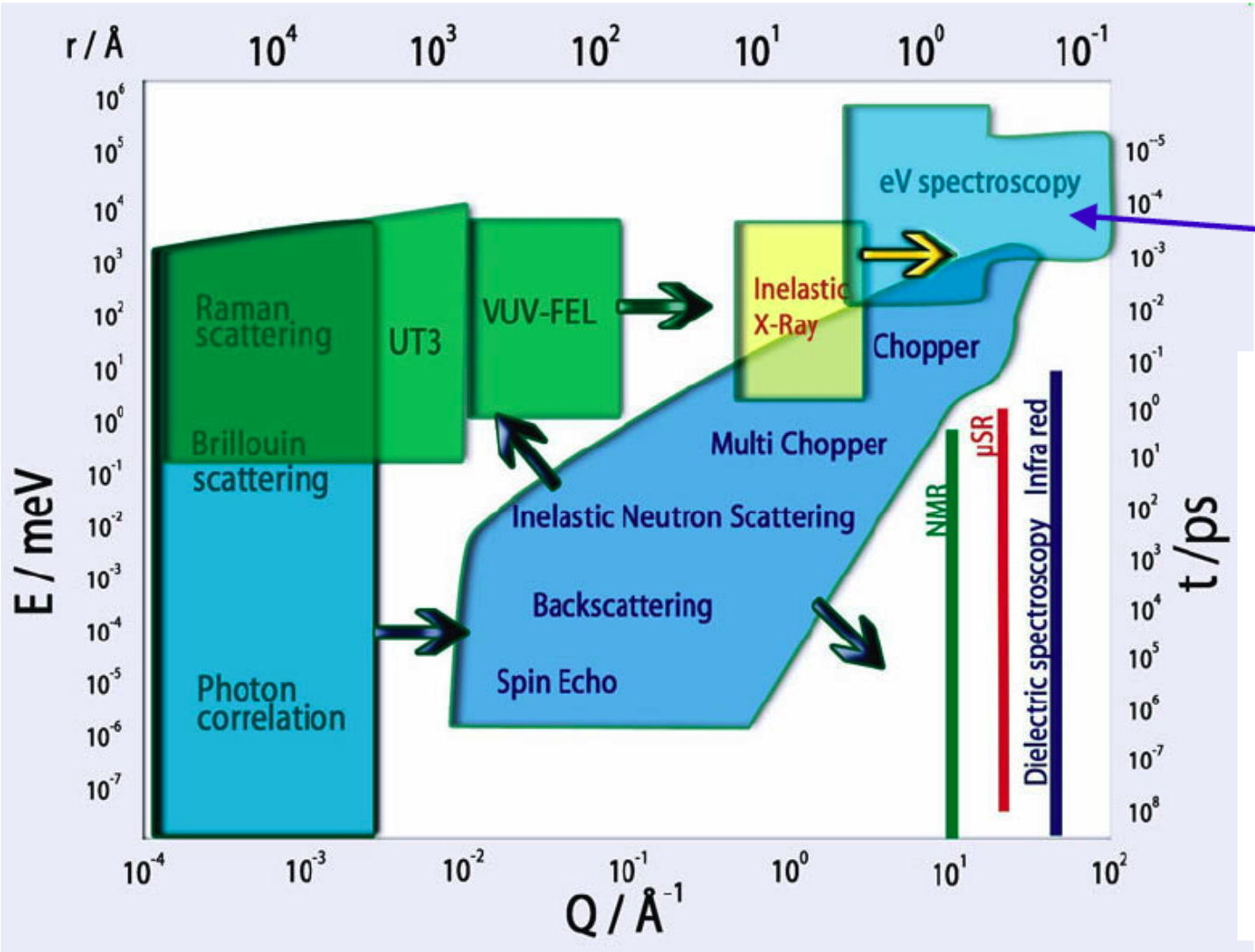
- **No choppers**
  - Neutrons are too fast
- **No crystal analysers**
  - Neutrons have too short λ

  Then …
- **Nuclear resonances**
  - Several experimental configurations since 1986……



$$t, s \rightarrow 0;\ s = \frac{\hbar \boldsymbol{Q}}{M}\, t$$

Struck Atoms travel only a short time- small distance



implying large

$$\boldsymbol{Q}, \omega$$

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

$\omega > 1\text{eV}$

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

$0.1\ \text{Å} < r < 0.2\ \text{Å}$

DINS (**D**eep **I**nelastic **N**eutron **S**cattering)

For a monoatomic system:

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{1}{\hbar}\frac{k_f}{k_i}\left[\frac{\sigma_c}{4\pi}S(\mathbf{q},\omega) + \frac{\sigma_i}{4\pi}S_i(\mathbf{q},\omega)\right]$$

Impulse Approximation regime : high q and ω

Initial state

$$S_{IA}(\mathbf{q},\omega) = \int n(\mathbf{p})\delta\left(\hbar\omega - \frac{\hbar^2 q^2}{2M} - \frac{\hbar\mathbf{q}\cdot\mathbf{p}}{M}\right)d\mathbf{p}$$

2. DINS

y is the West scaling variable:

$$y = \boldsymbol{p} \cdot \hat{\boldsymbol{q}} = \frac{\mathrm{M}}{\hbar^2 q}\left(\hbar\omega - \frac{\hbar^2 q^2}{2\mathrm{M}}\right)$$

recoil energy

or longitudinal momentum distribution

$$S(q,\omega) = \frac{\mathrm{M}}{\hbar q}\mathrm{J}(\mathrm{y})$$

NCP

3. DINS

At finite q - Final State Effects (FSE)

Interactions among recoiling particle and the Surroundings → inter- and intramolecular interactions:

- Response function is q dependent --->F(y,q)
- At high q dominant effect comes from the intramolecular interactions

Where and How do we make the measurement

…

Effective thermal neutron flux n/cm²-s
10^20
10^15
10^10
10^5
1
1930
1940
1950
1960
1970
1980
1990
2000
2010
Chadwick
350 mCi
Ra-Be source
Berkeley 37-inch cyclotron
CP-1
CP-2
X-10
NRX
MTR
NRU
HFIR
HFBR
ILL
ZINP-P/
WNR
LINP-P
IPNS
KENS
ISIS
SNS
Reattori
Sorgenti Pulsate

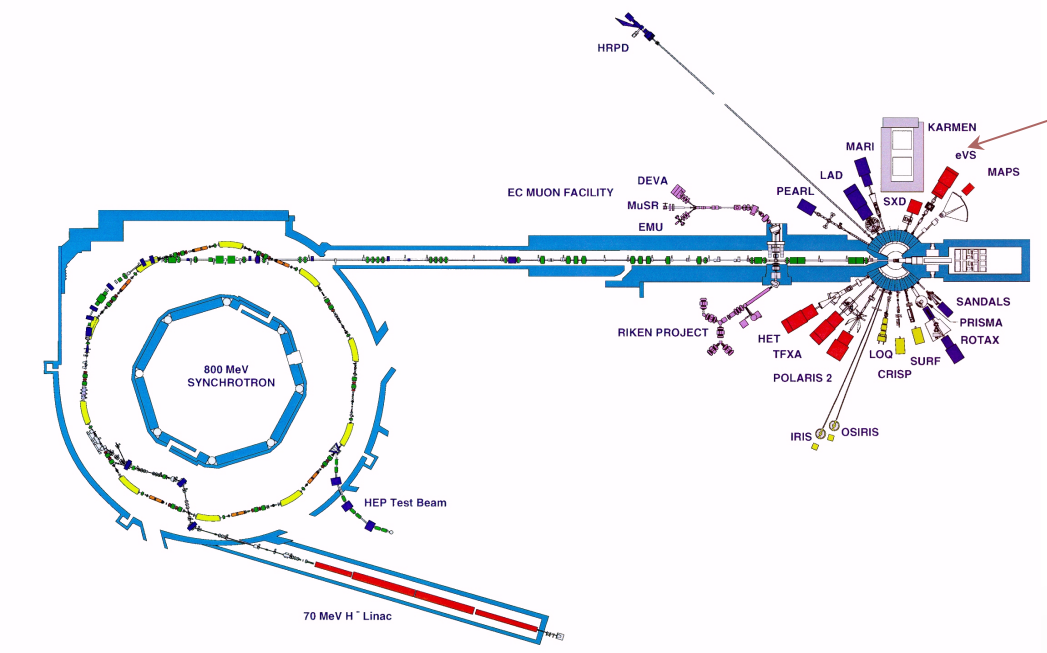
Single atom short-scale dynamics

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†‖



Vesuvio is here!

Inelastic scattering with eV neutrons

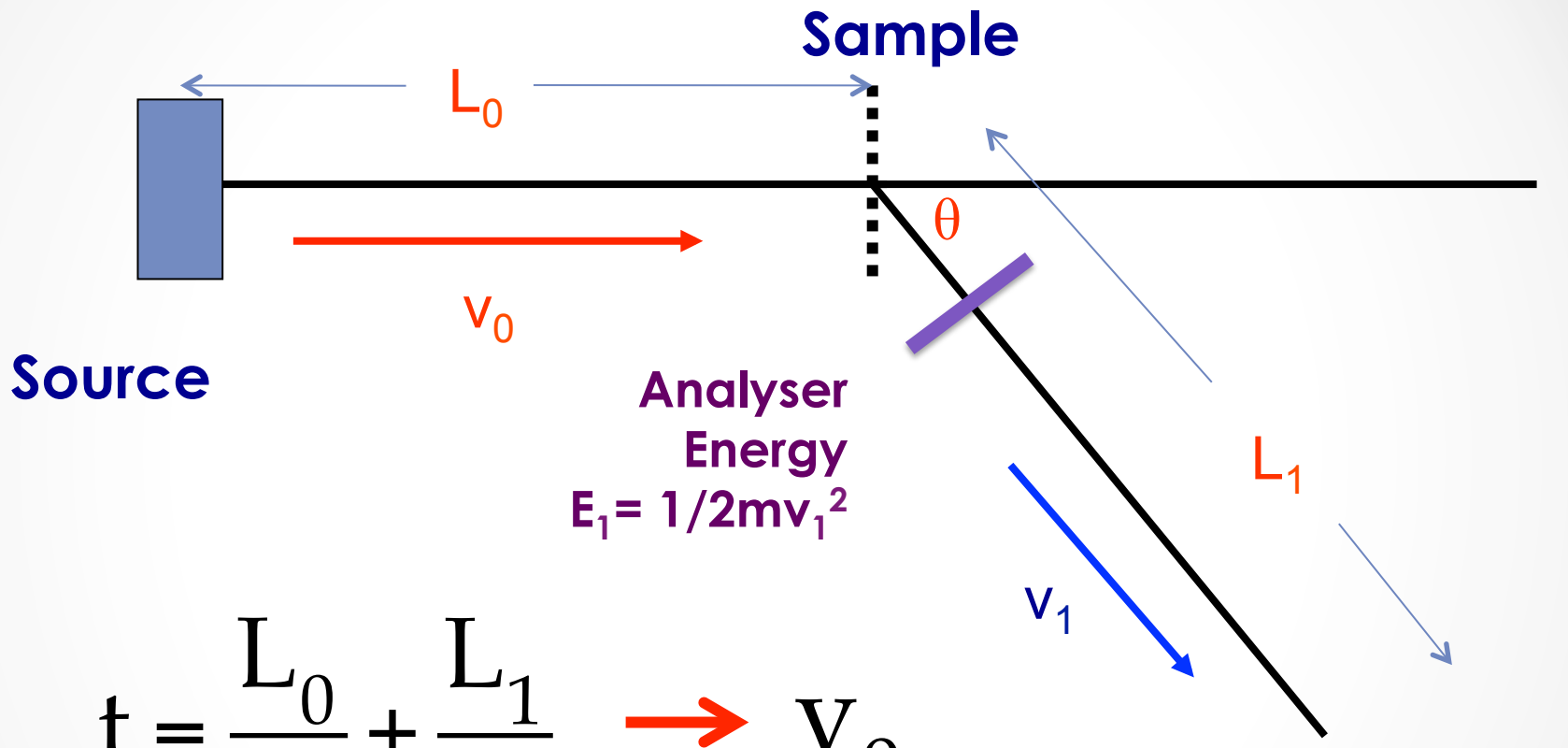
The Resonance Filter Spectrometer (1986-2002)

- Filter Difference technique (FD)
- Double Difference Filter technique (FDD)

Resonance Detector Spectrometer (from 2002)

- Foil cycling technique (FC)

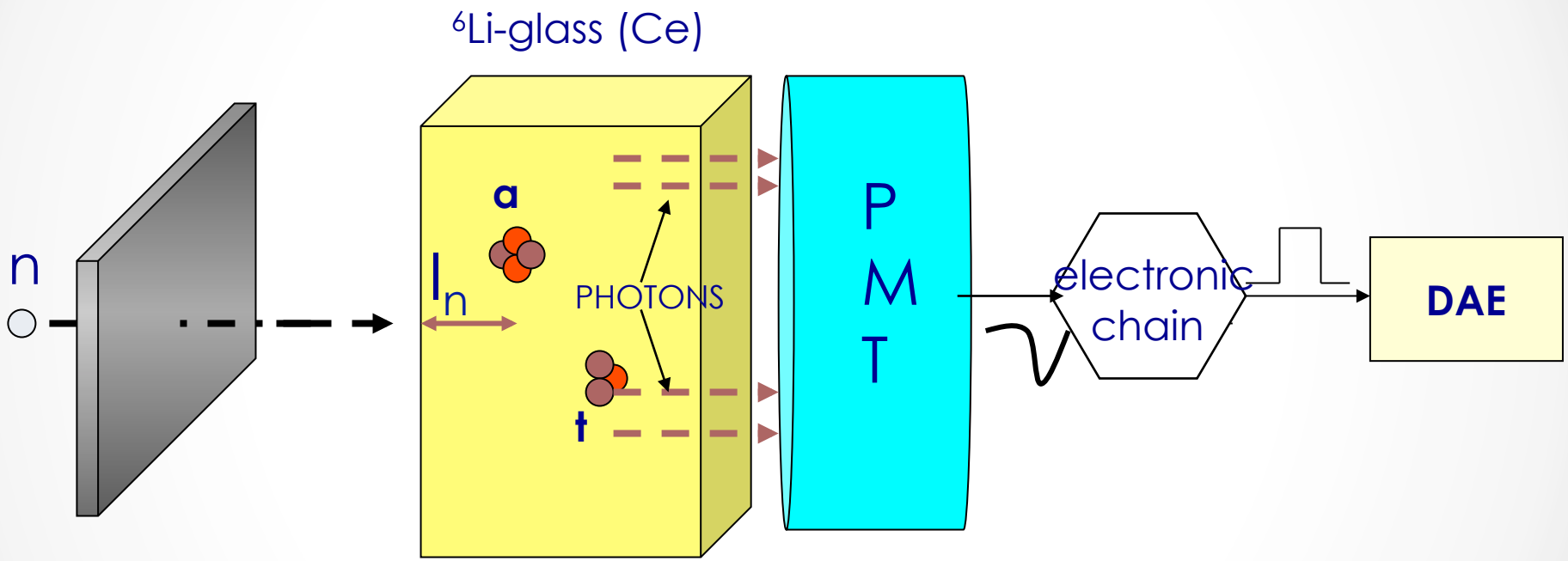
Time of flight measurements at eV



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

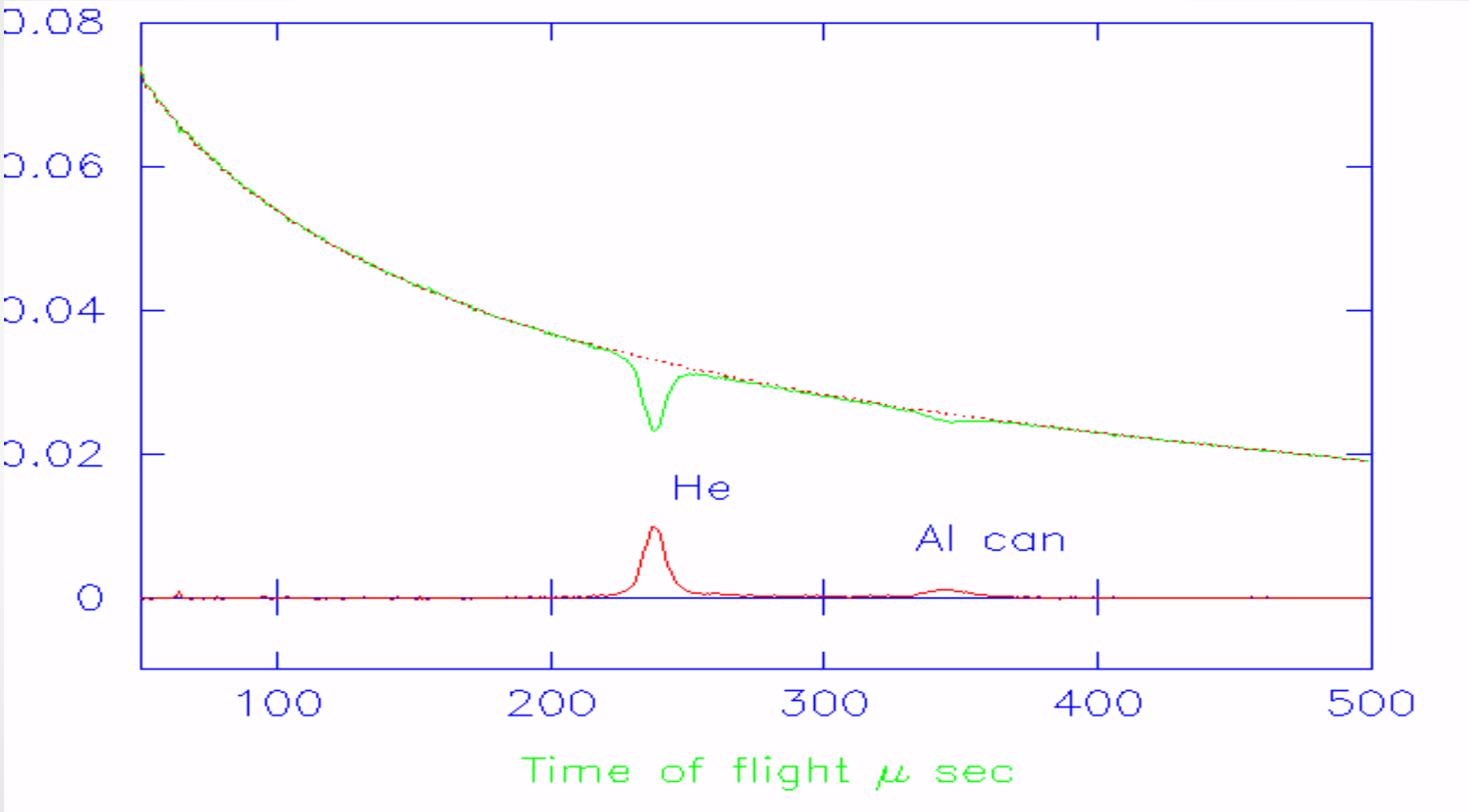
Filter Difference Technique

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



$$\eta_{\text{Li-glass}} \cong 1/v$$

The Filter Difference Technique on eVS Spectrometer at ISIS



J. M.F.Gunn et al. "A new approach to impulsive neutron scattering" Journal of Physics C 19, L835 (1986)

Count rate as a function of *t*

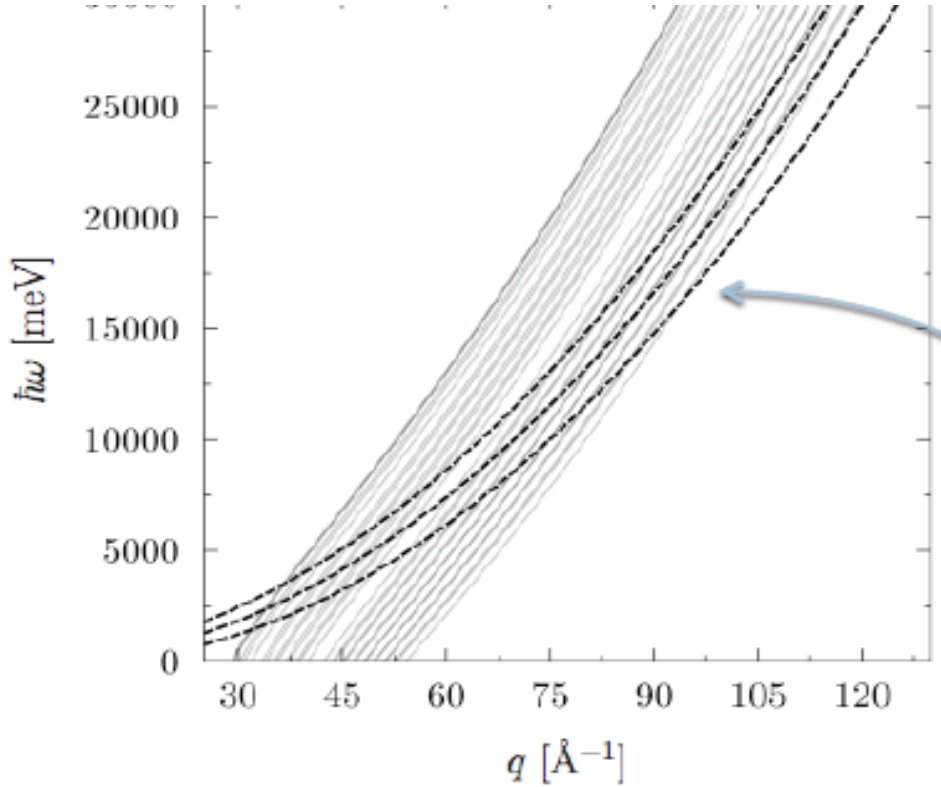
$$C(t) = 2\left(\frac{2}{m}\right)^{1/2} \frac{E_0^{3/2}}{L_0} I(E_0) D(E_1) \sum_M N_M \frac{d^2\sigma_M}{d\Omega dE_1} d\Omega$$

probability that a neutron of energy E_1 is detected

$$\frac{d^2\sigma_M}{d\Omega dE_1} = b_M^2 \sqrt{\frac{E_1}{E_0}} \frac{M}{q} J_M(y_M)$$

$$y_M = \frac{M}{q}\left(\omega - \frac{q^2}{2M}\right)$$

- In the IA q and ω are no longer independent variables
- Detectors at all angles give the same information for isotropic samples



Inelastic neutron scattering at High energy-high momentum transfer

Probe a particular excitation:

$$\hbar\omega = \frac{\hbar^2 q^2}{2M}$$

$$S(\vec{q},\omega) = \hbar \int d\vec{p}\, n(\vec{p})\, \delta\left(\hbar\omega - \frac{\hbar^2 q^2}{2M} - \frac{\vec{p}\cdot\hbar\vec{q}}{M}\right)$$

Vesuvio Beamline at the ISIS neutron source -UK

CMCSN

Any scan in q, ω space which crosses the line $\omega = q^2/(2M)$ gives the same information in isotropic sample

$$\Delta y^2 = \sum_i [(\partial y/\partial \omega)(\partial \omega/\partial x_i) + (\partial y/\partial q)(\partial q/\partial x_i)]^2 \, \Delta x_i^2$$
$$= \sum_i [M/q(\partial \omega/\partial x_i) - (\partial q/\partial x_i)]^2 \, \Delta x_i^2 .$$

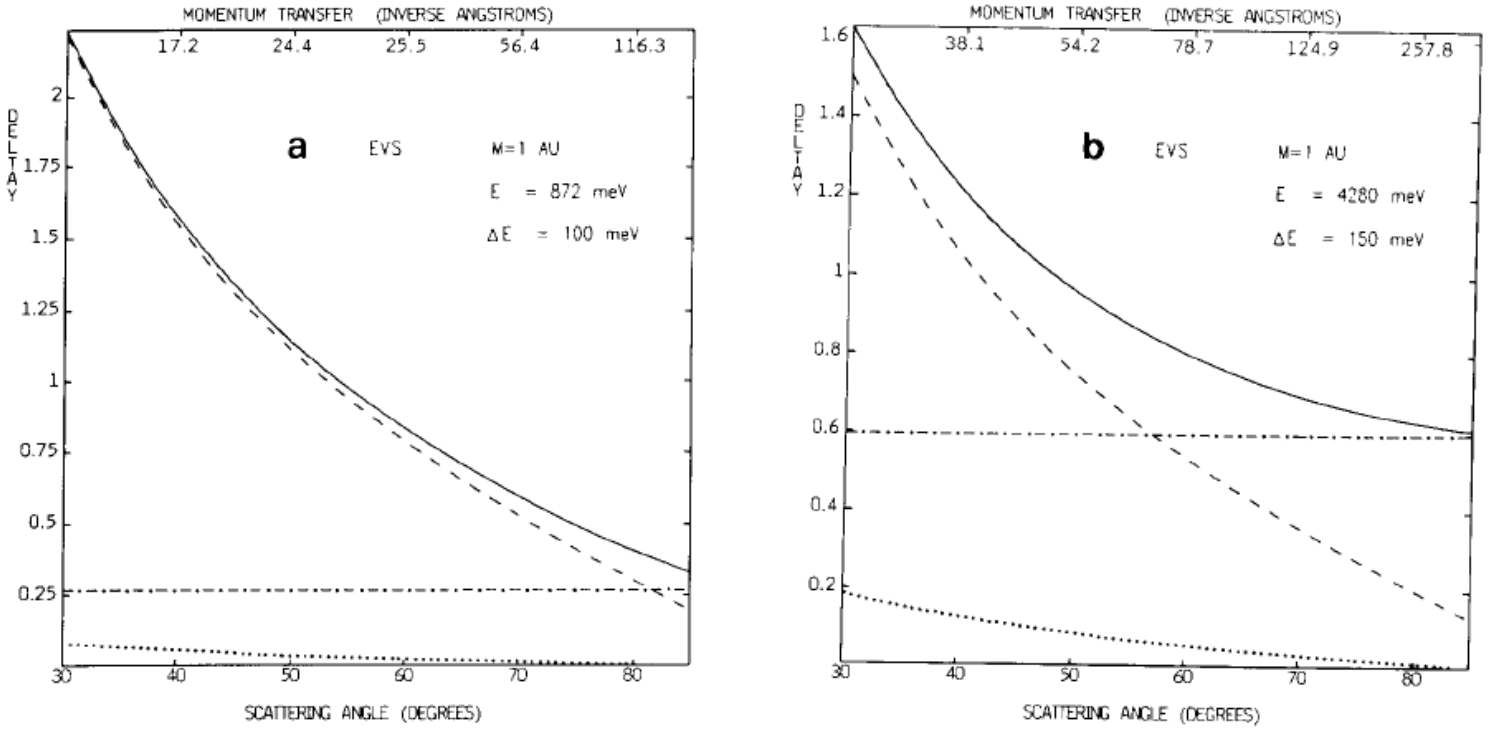


Fig. 2. The total resolution (———) and the energy (— — —), angular (·–·–·) and timing (·····) contributions to the resolution in hydrogen ($M = 1$ amu) shown as a function of scattering angle and momentum transfer for eVS at (a) $E_1 = 872$ meV and (b) $E_1 = 4280$ meV.

C. Andreani, G. Baciocco, J. Mayers, R. Holt, Nuclear Instruments and Methods A276, 297 (1989),

RESOLUTION COMPONENTS

Geometrical ↦ Gaussian

Energy ↦ Gaussian & Lorentzian

Single Difference (FD)

U resonances:

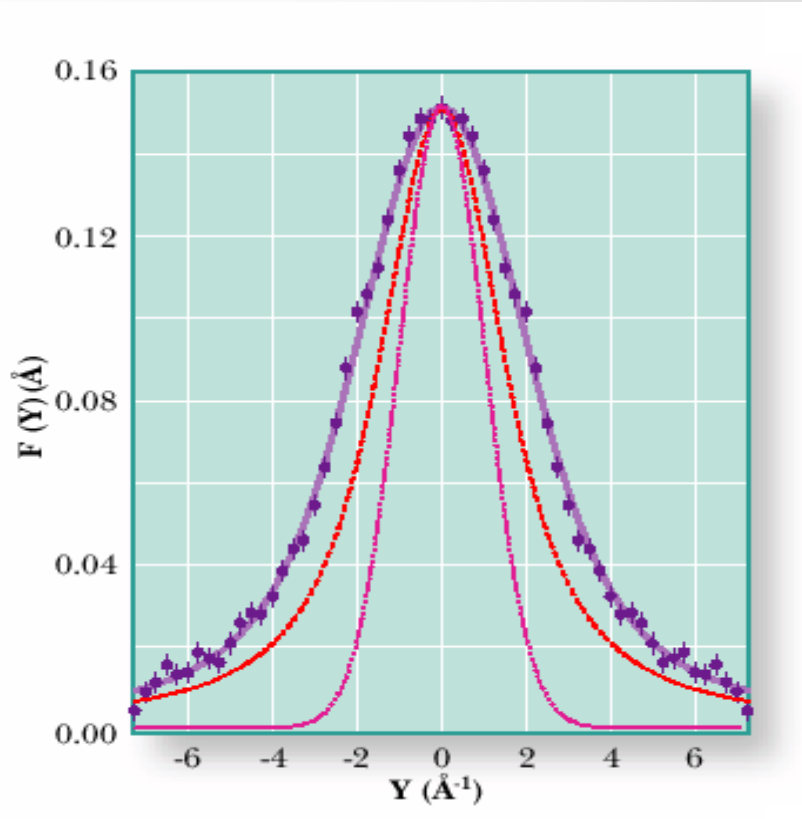
↦ 6.7eV, 20.7eV, 37eV..

FWHM (at 6.7 eV) ↦ 0.04 eV

Doppler broadning

at RT ↦ 0.11 eV

at 70 K ↦ 0.06 eV



Scattering function F(y) for the ^{3}He bcc solid sample. Data (full circles); best fit (purple line); resolution function FD (red line),

DOUBLE DIFFERENCE TECHNIQUE

$$R_2(E) = [1 - T_1(E)] + \frac{t_1}{t_2}[1 - T_2(E)]$$

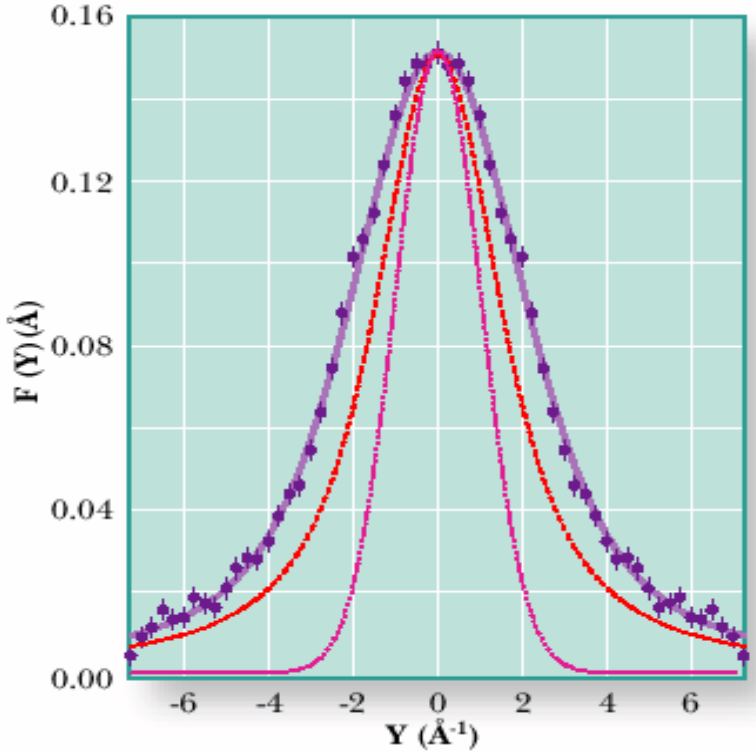
$$1 - T_1(E) = 1 - \exp[-Nt_1\sigma(E)] \sim Nt_1\sigma(E)$$

$$1 - T_2(E) = 1 - \exp[-Nt_2\sigma(E)] \sim Nt_2\sigma(E)$$

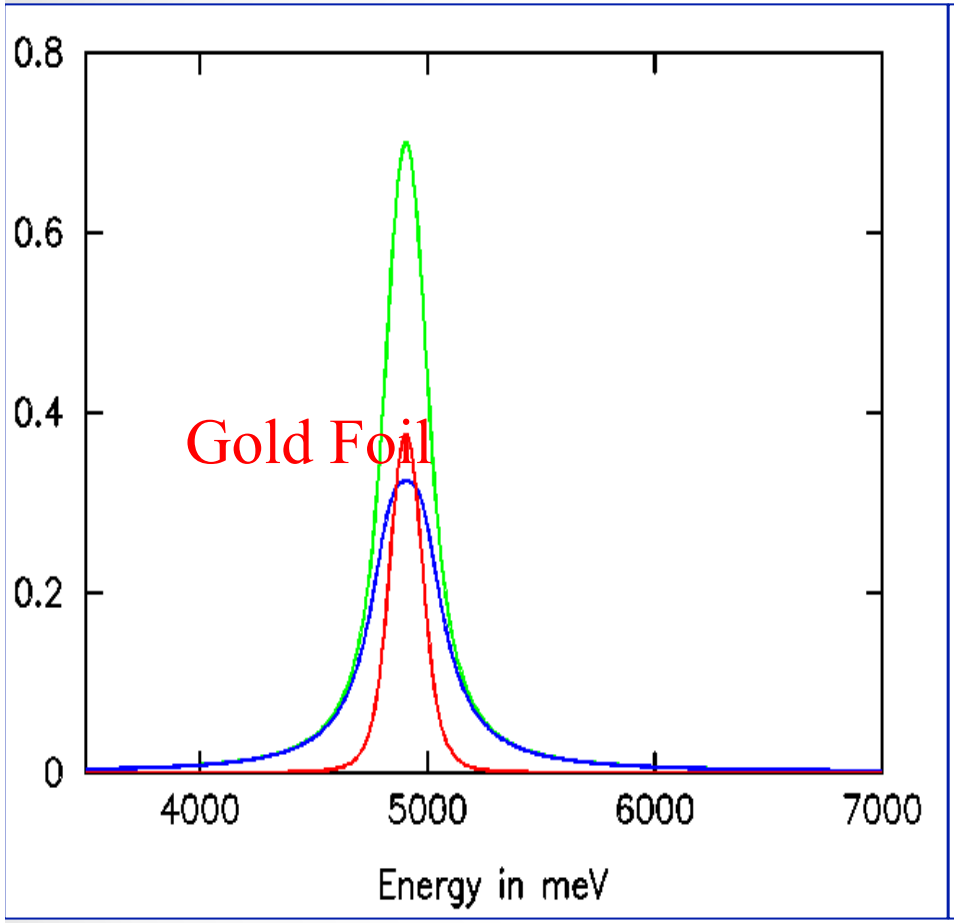
When $\sigma(E)$ is small
Lorentzian wings are removed

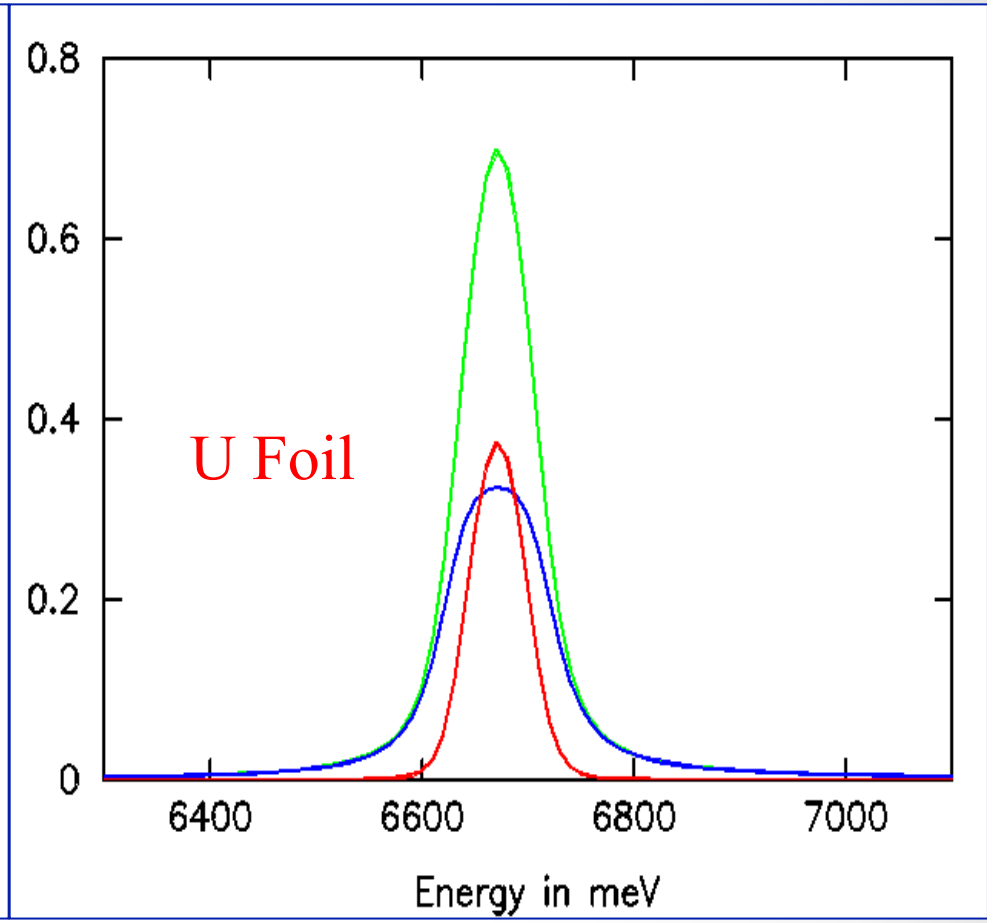
★ Resolution reduction of ~2 for U and Au foils

Scattering function F(y) for the ^{3}He bcc solid sample. Data (full circles); best fit (purple line); resolution function FD (red line); simulated resolution function FDD for VESUVIO (line).



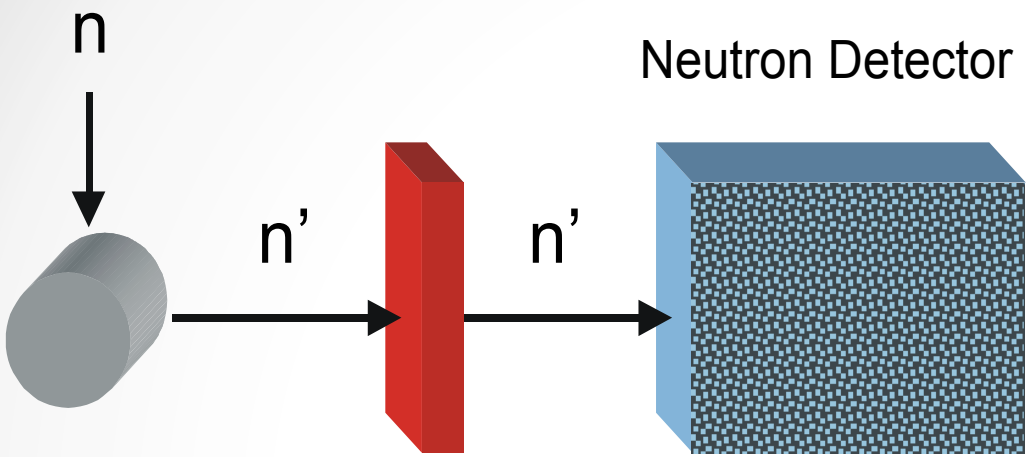
DOUBLE DIFFERENCE TECHNIQUE



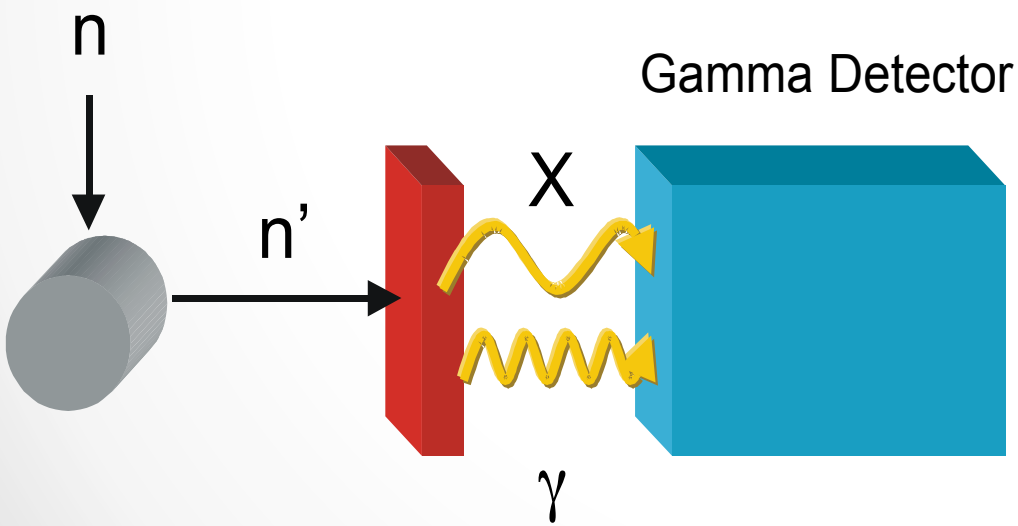


$$1-e^{-Nt\sigma}=Nt\sigma \quad \text{as } \sigma \rightarrow 0$$

The Resonance Detector Spectrometer, 2002

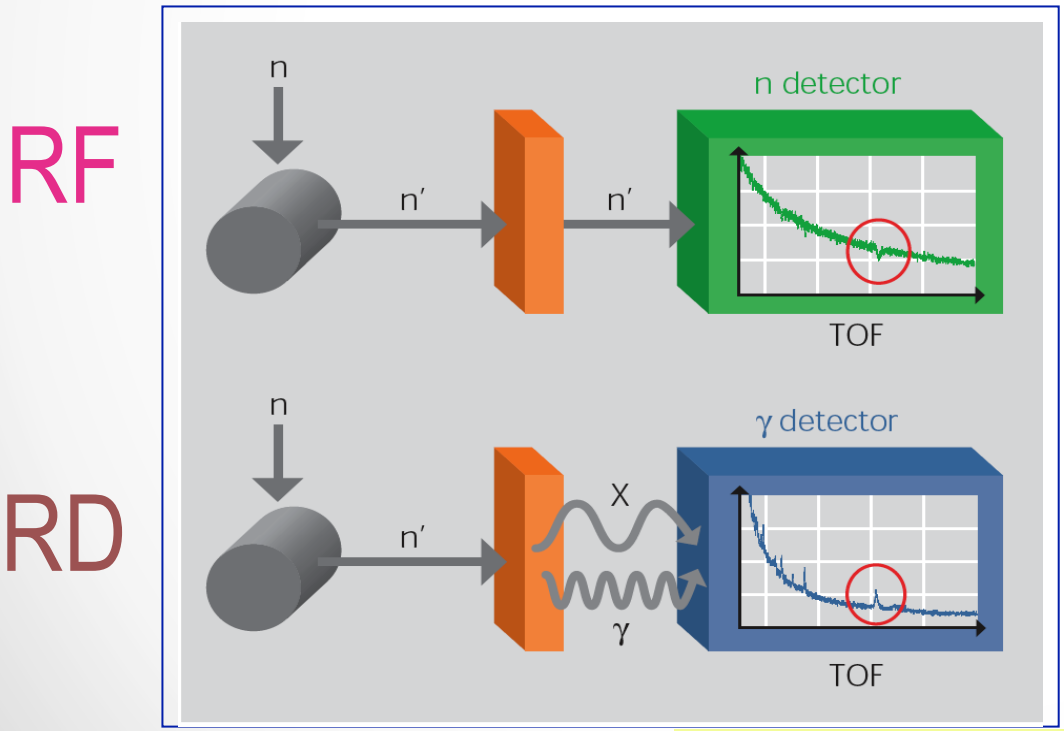


Resonance Filter Spectrometer



Resonance Detector Spectrometer: a photon counter detects the γ and X-ray emission following γ resonant neutron absorption in a converter foil

Principles of Resonance Detector (RD) techniques

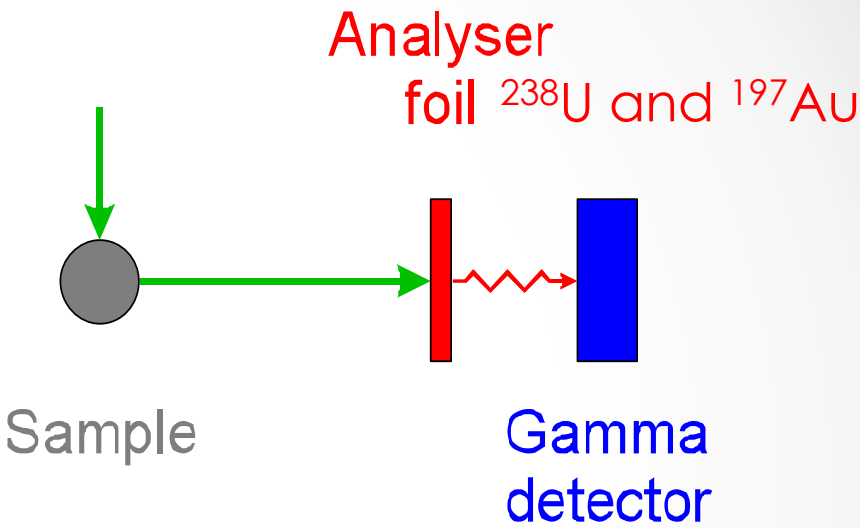


YAP scintillator

The Resonance Detector Spectrometer, 2002

Gamma detector, why?:

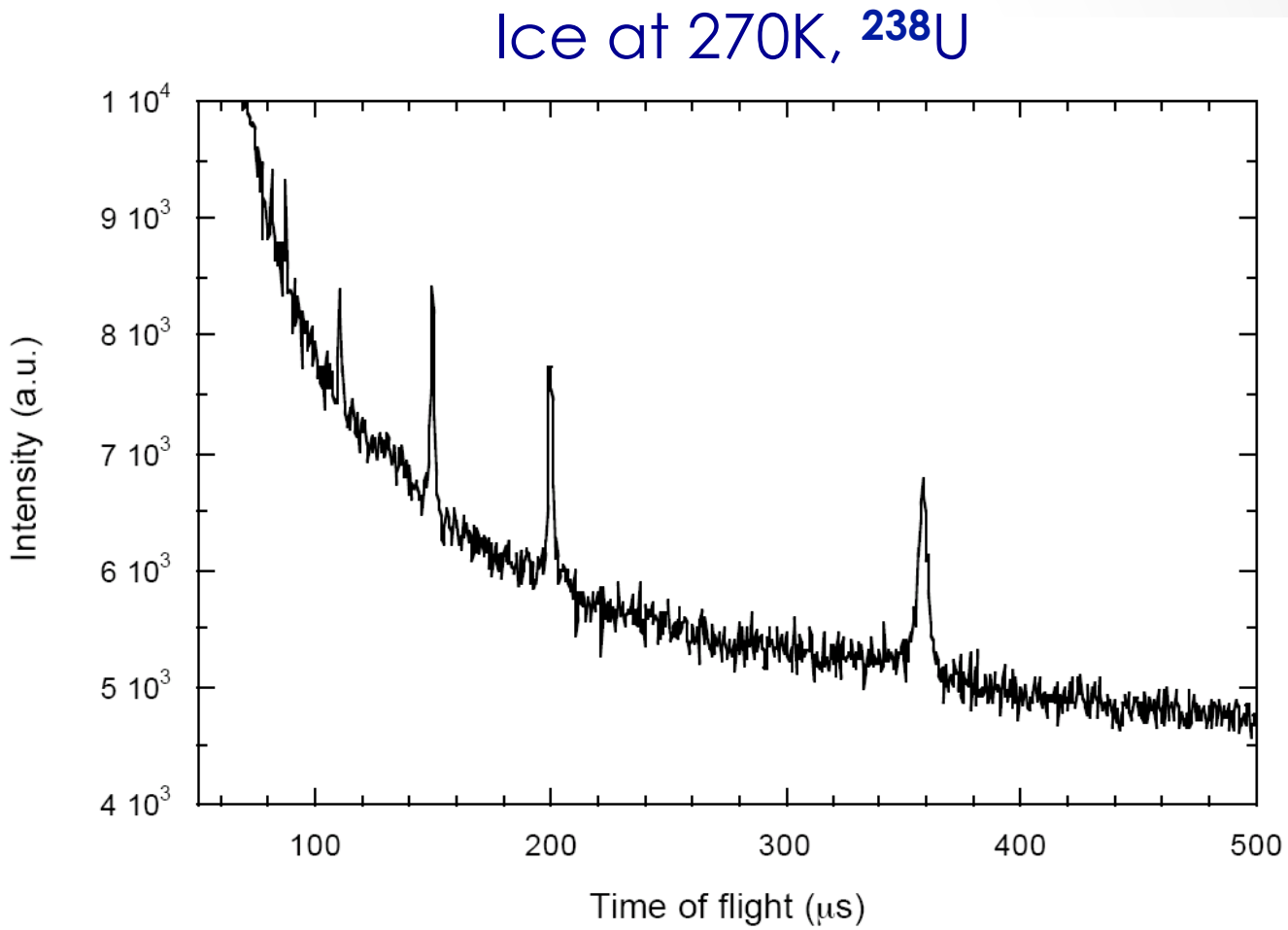
- Neutron detector very inefficient at ~ 100 eV
- Gamma detector efficiency independent of neutron energy

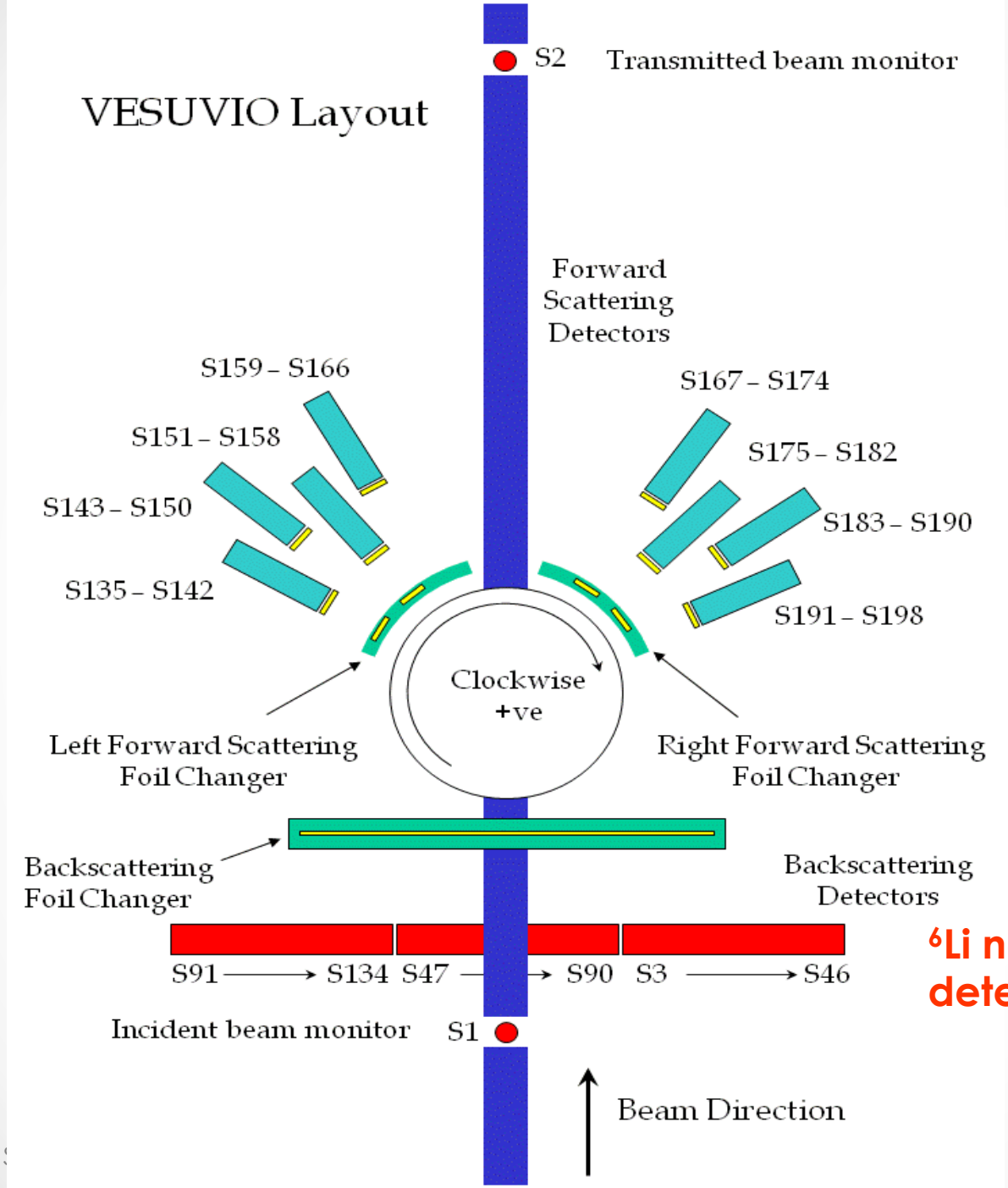


Experimental Tests at ISIS on YAP

The Resonance Detector Spectrometer, 2002

- YAP detector at 2°

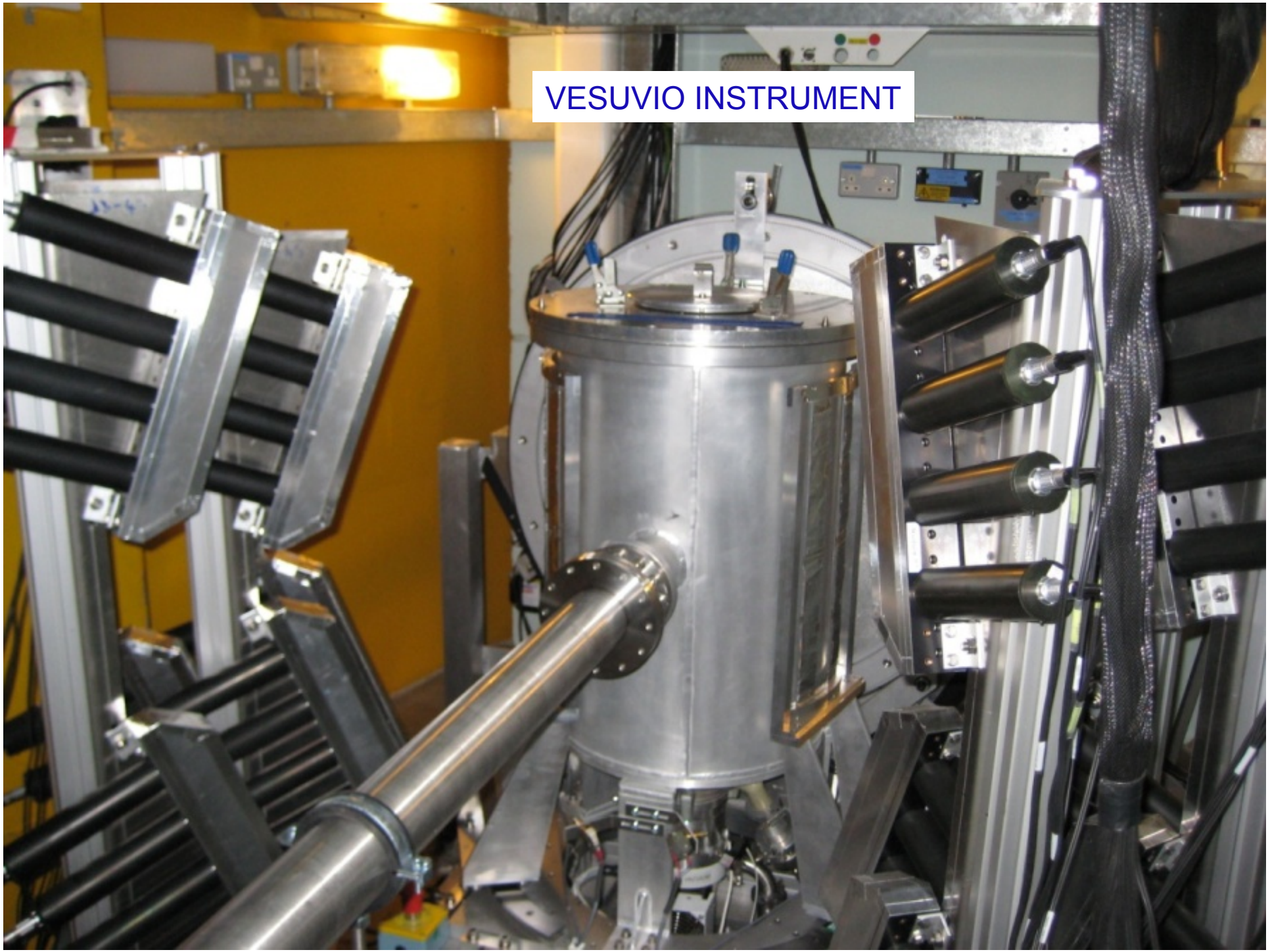




2006

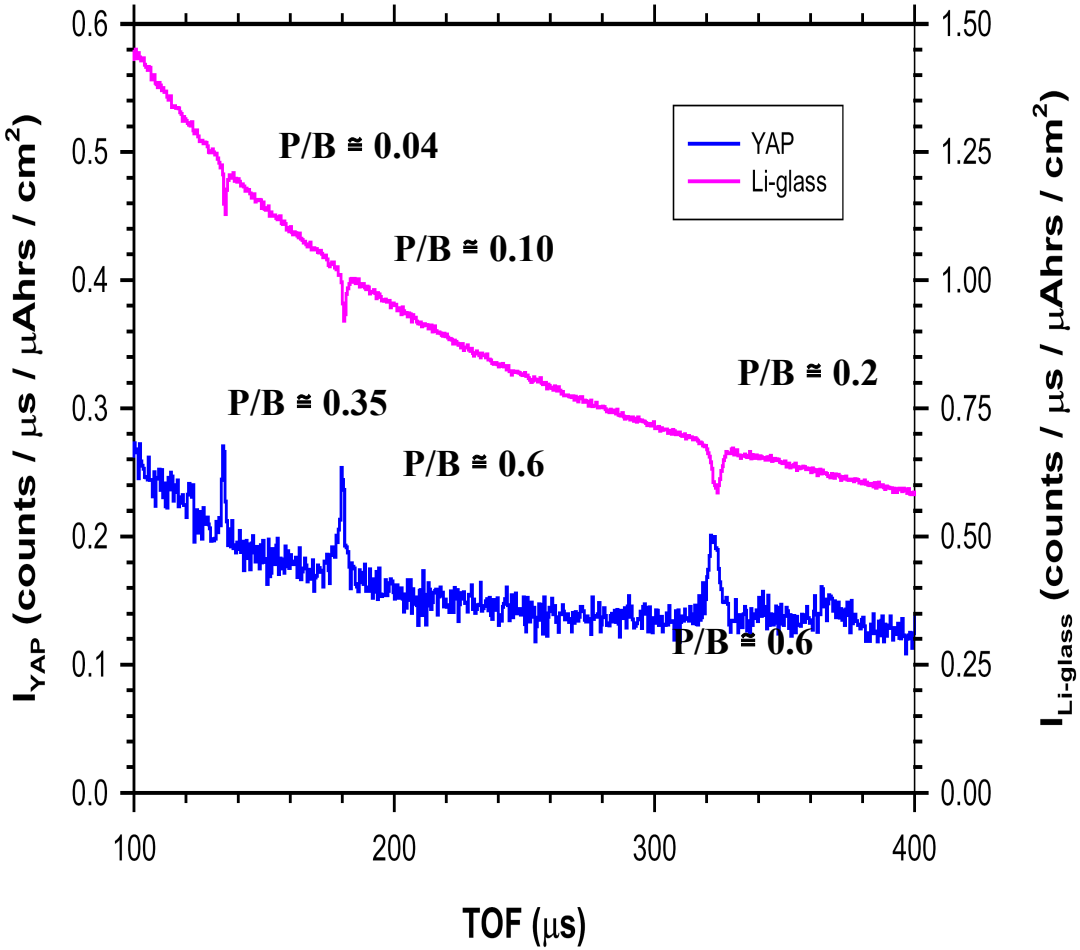
YAP gamma detectors

^{6}Li neutron detectors


VESUVIO INSTRUMENT

YAP: a comparison with Li-glass

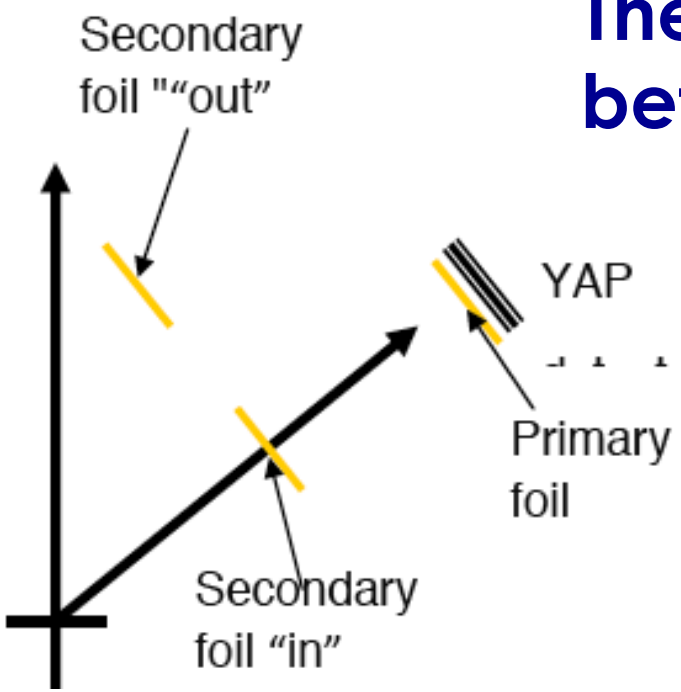
YAP has much better Peak to Background (P/B) ratio as compared to Li-glass



YAP detectors give

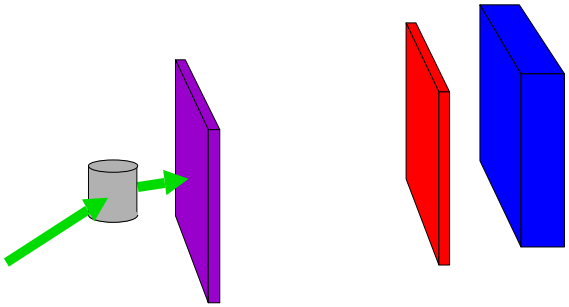
- Energy Resolution FWHM reduced by ~2
- Better resolution peak shape
- 100 times less counts on filter in and filter out measurements…thus less detector saturation at short times
- Similar count rates in the differenced spectra
- Larger differences between foil in and foil out measurements….therefore more stability over time.
- Order of magnitude improvement in measurement accuracy

The "raw" data is the difference between foil out and foil in data.



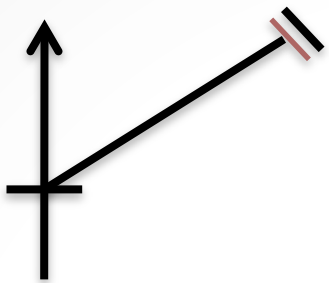
The cycling removes drifts in detector efficiency with time, due for example to ambient temperature changes.

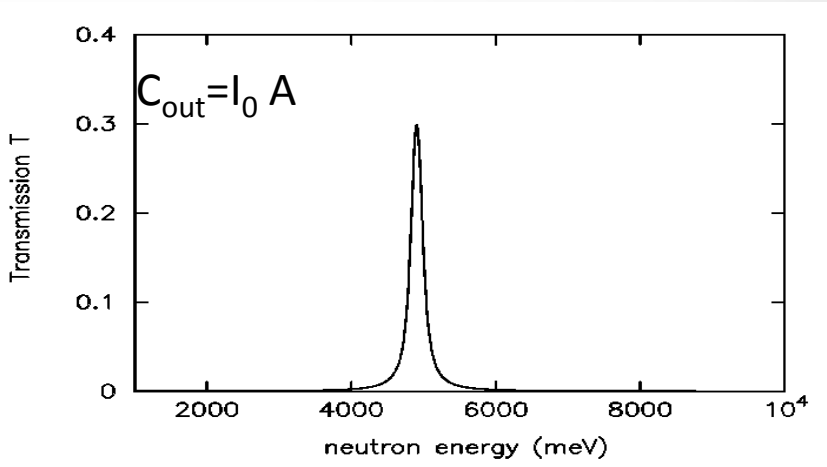
Experimental Tests at ISIS in 2005



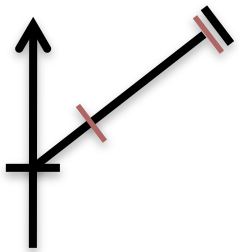
E M Schoonveld, J. Mayers et al Rev. Sci. Inst. 77 95103 (2006)

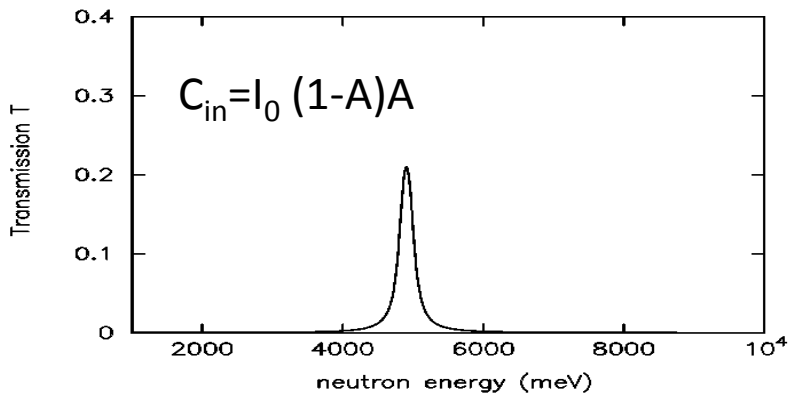
Foil out



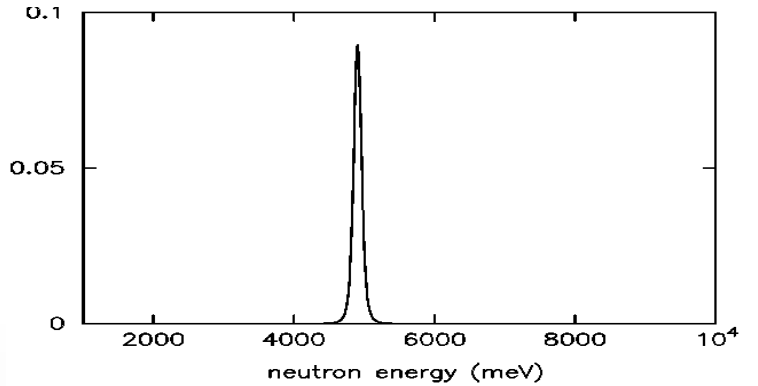


Foil in





$$C=C_{out}-C_{in}= I_1\ [\ 1-A^2]$$



$$C_{out}(E_0, E_1, \theta) = C(E_0, E_1, \theta) A(E_1) \eta + B_{out}(E_0, E_1, \theta)$$

Foil out

Rate at which scattered neutrons are incident on the detector after scattering by the sample.

Background with the secondary foil in the "out" position.

Probability that the ray cascade is registered by YAP

Probability that a neutron of energy E_1 is absorbed in the primary foil.

$$C_{in}(E_0, E_1, \theta) = C(E_0, E_1, \theta)[1 - A(E_1)] A(E_1) \eta + B_{in}(E_0, E_1, \theta)$$

Foil in

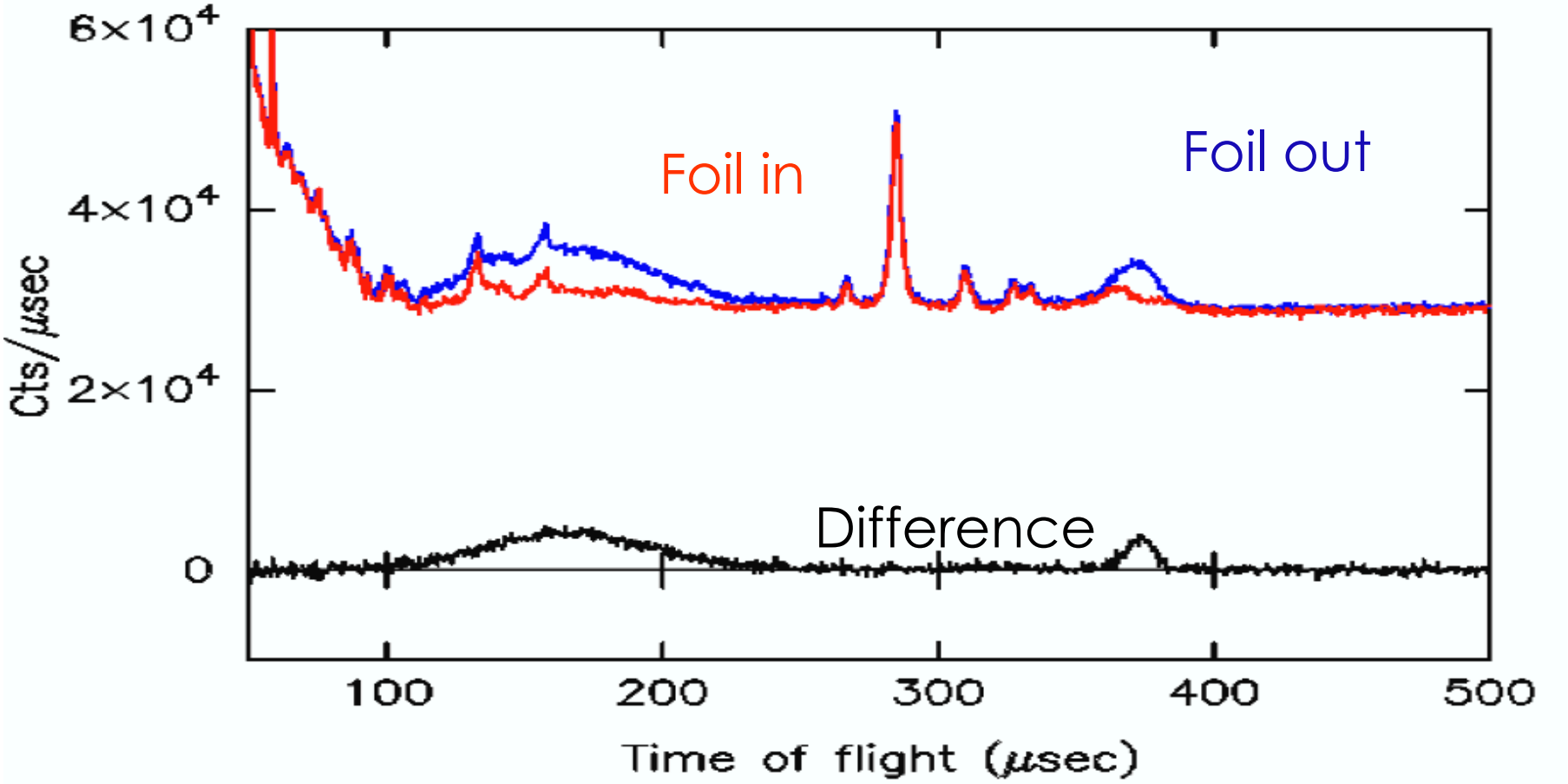
Counts from the primary foil are reduced by this factor

Background with the secondary foil in the "in" position.

$$(C_{out} - C_{in}) = C \eta A^2 + (B_{out} - B_{in})$$

VESUVIO RAW DATA

Foil cycling technique

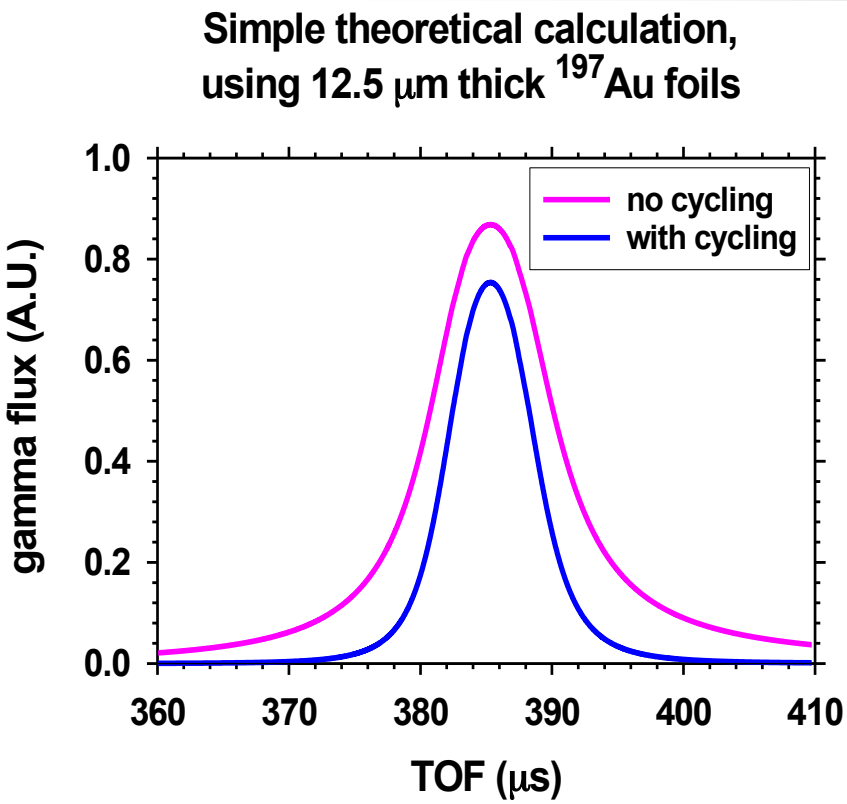


ZrH_2 $2\theta = 67$

Total Count Time = 5600 μ-A-hours
Total neutron pulses = 7.82×10^6

Foil cycling technique

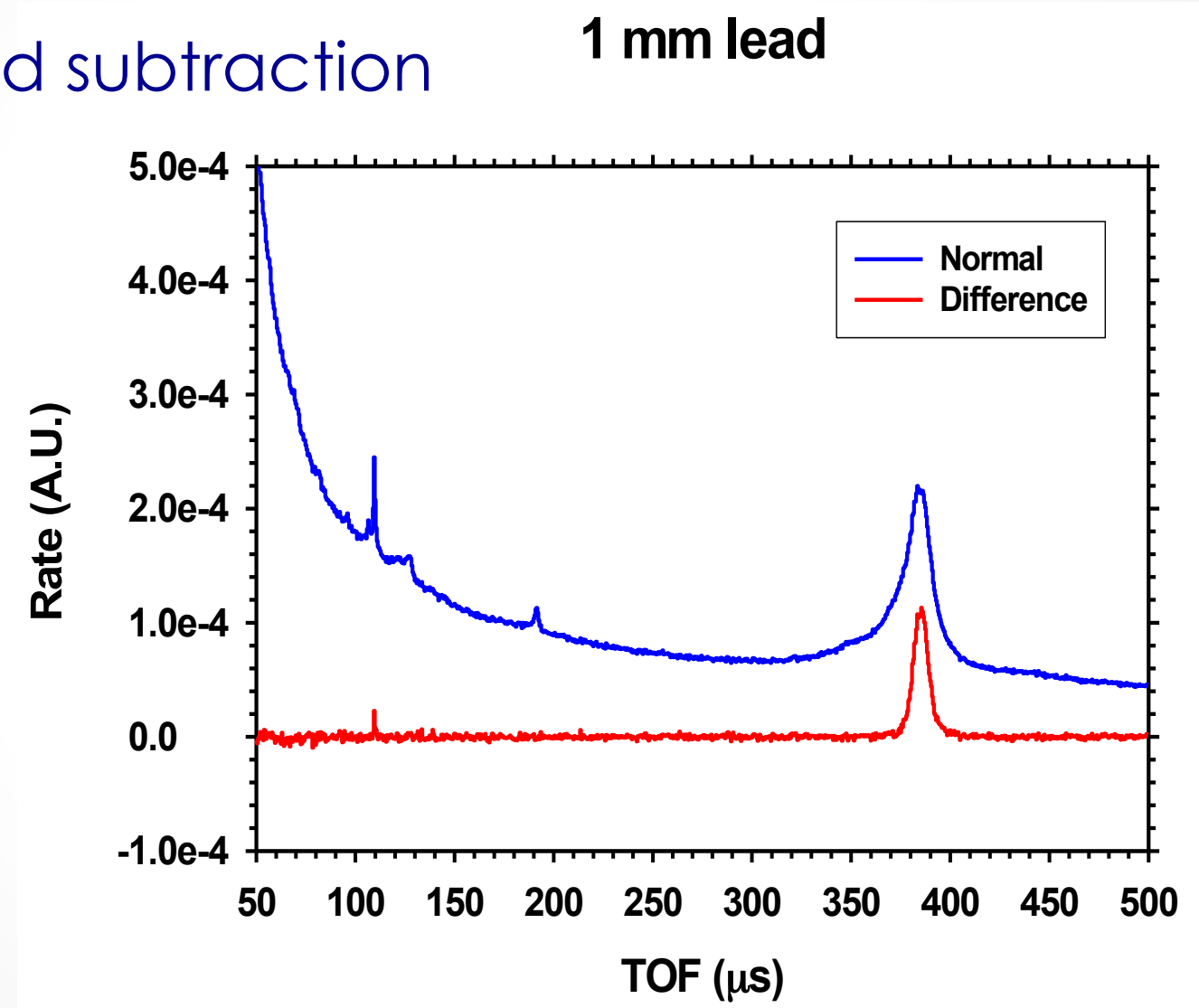
- Resolution improvement
- Same thickness as analyser foil:
  Background subtraction + resolution improvement



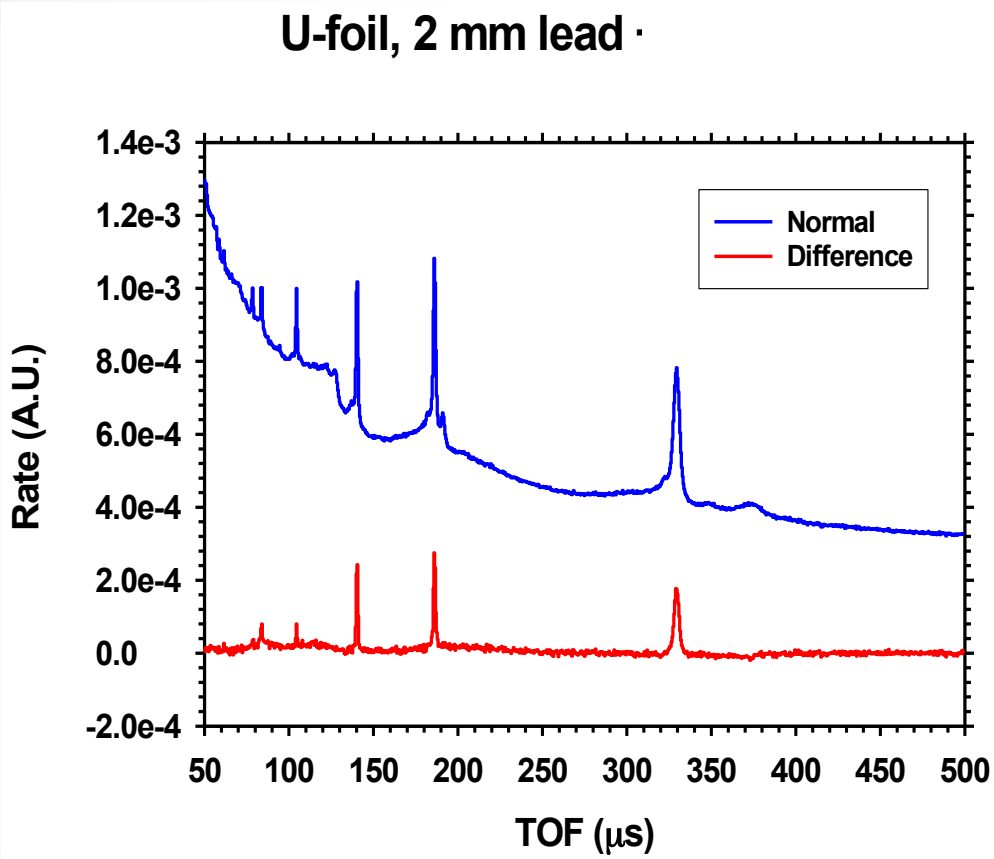
- **Narrower peak**
- **Considerably smaller tails**
- **Don't loose too much efficiency**
- **Even narrower peak with thinner cycling foil, but loose efficiency**

Measurements on VESUVIO, 2005

Background subtraction



Measurements on VESUVIO, 2005

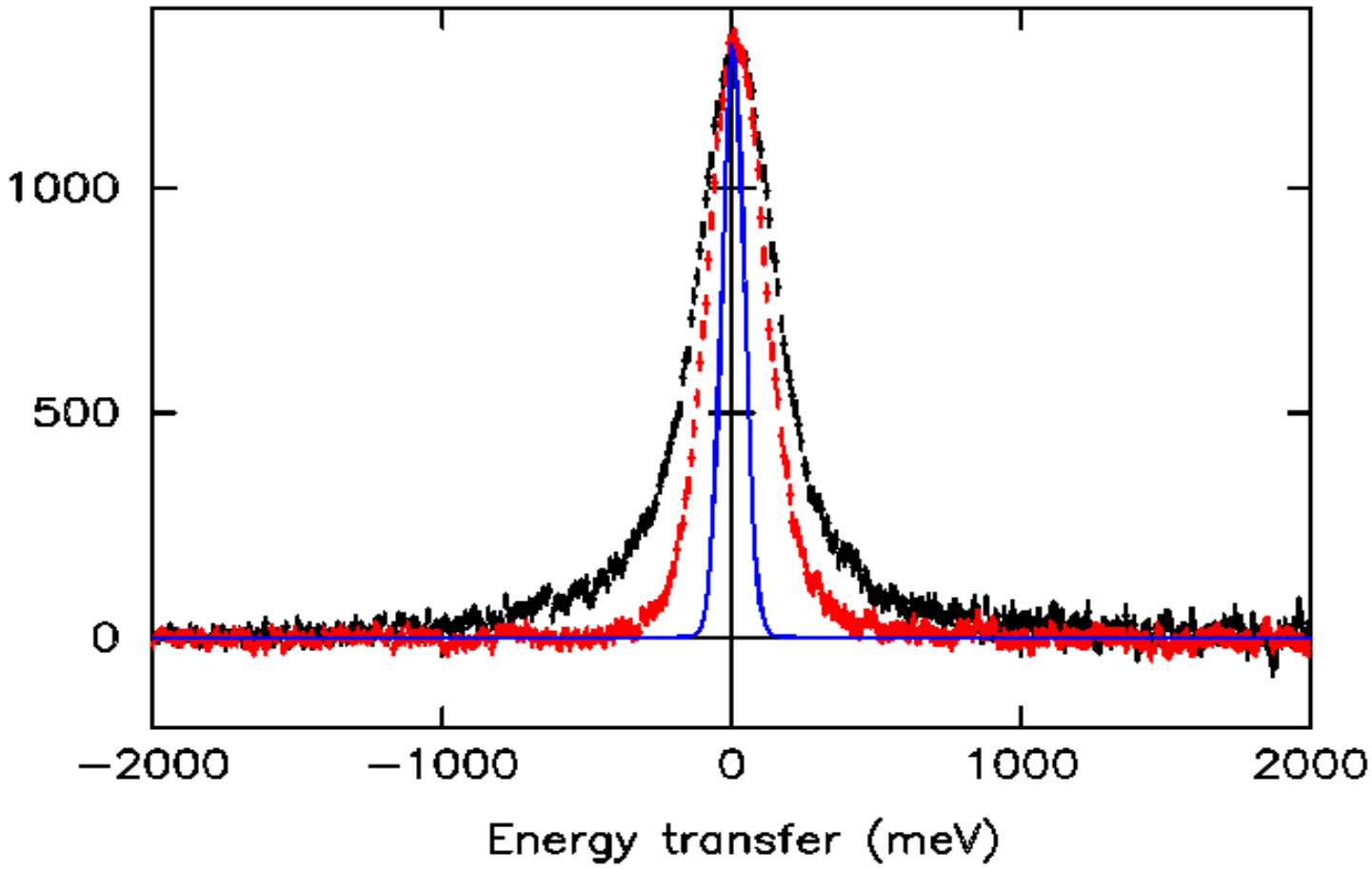


All 3 main resonance peaks usable

Need thicker foils for higher energy resonances

Background subtraction works very well

Resolution improvement FC versus FD

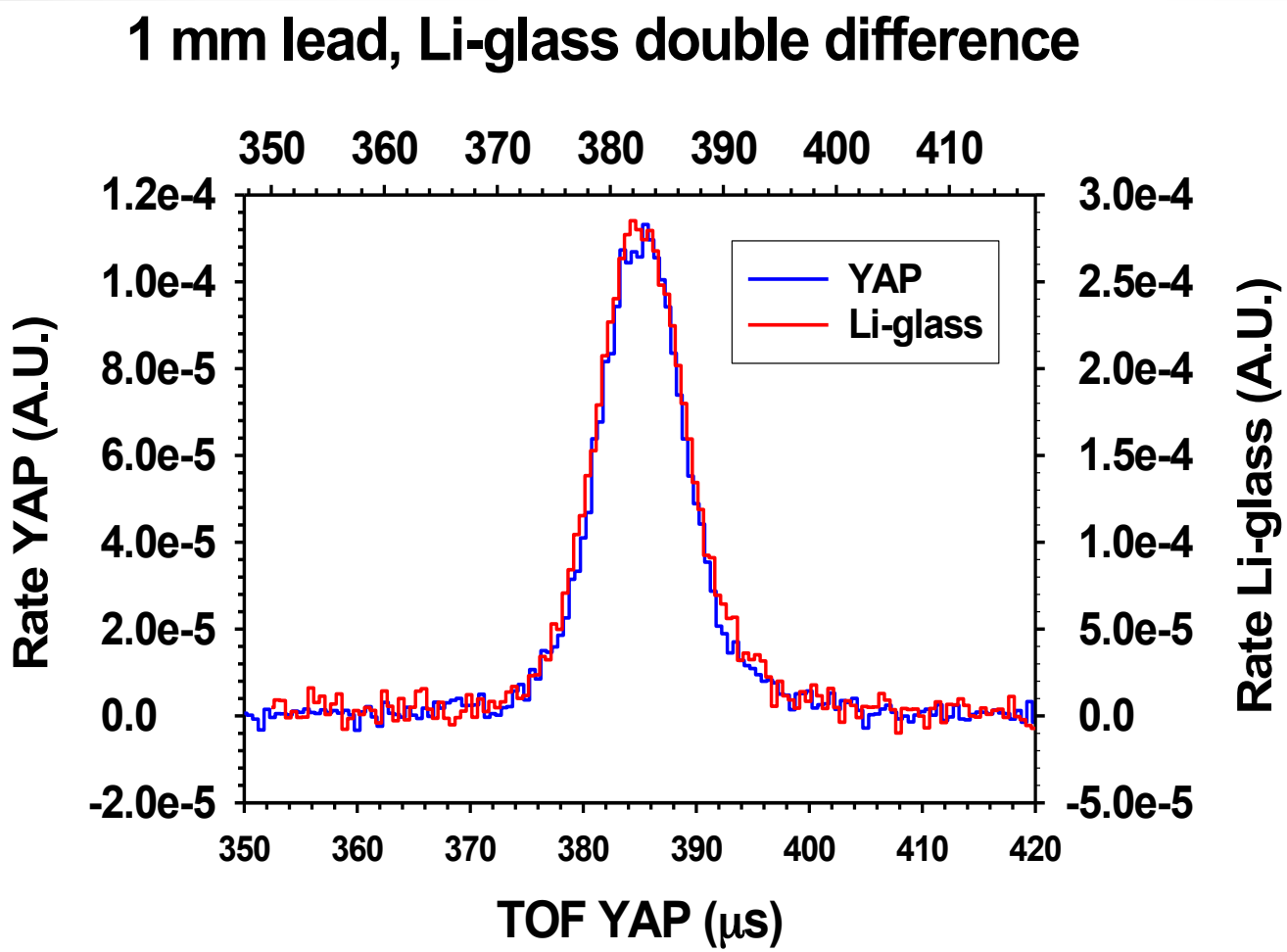


Blue = intrinsic width of lead peak

Black = measurement using Filter Single Difference technique

Red = Foil Cycling technique

Resolution improvement FC versus FDD



FWHM=9.5 μs

FWHM = 9.2 μs

YAP same resolution as Li-glass in double difference

Summary

- Current VESUVIO has order of magnitude better accuracy for proton measurements than 3 years ago due to installation of YAP detectors
- Current measurement accuracy of widths of proton momentum distributions is ~0.5%.

Future Developments

- Gamma background reduction by moving shielding further from instrument
- Residual background could be virtually eliminated by circular geometry for forward scattering detectors: Need detectors on rings
- Rotate secondary foils keeping the foil scattering angle constant should almost eliminate gamma background effects

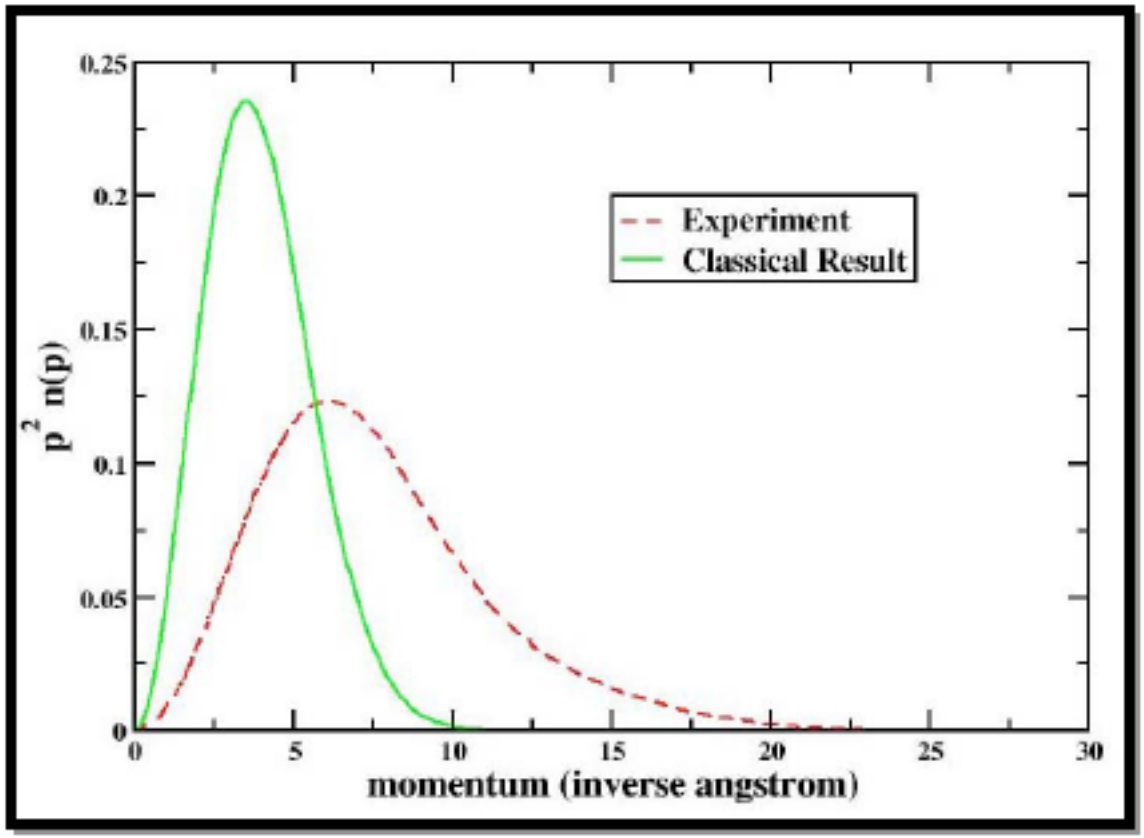
Short-time single particle dynamics

↓

nuclear quantum effects

Nuclear quantum effects

Not only the electrons but also the protons need quantum mechanical description



The Proton Momentum Distribution n(p) in water probed by Neutron Compton Scattering (NCS) displays importance of nuclear quantum effects

Courtesy of Roberto Car (Princeton University)

Nuclear quantum effects

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

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

→ **State of H in water dominated by ground state**

$\langle E_K \rangle$ & *n*(*p*) → PES

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

1. Classical systems

Classical case:

$$\mathrm{n}(p) = \frac{1}{\left(\sqrt{2\pi\,\sigma^2}\right)^3} \exp\left(-\frac{p^2}{2\,\sigma^2}\right)$$

With $\sigma^2 = \dfrac{M\,k_B\,T}{\hbar^2}$

2. Weak quantum systems

$$\mathrm{n}(p) = \frac{1}{\left(\sqrt{2\pi\,\sigma_{\mathrm{eff}}^2}\right)^3} \exp\left(-\frac{p^2}{2\,\sigma_{\mathrm{eff}}^2}\right)$$

$$\mathrm{T}_{\mathrm{eff}} = \mathrm{T} + \frac{\hbar^2}{12\,\mathrm{M}\,\mathrm{k}_{\mathrm{B}}\,\mathrm{T}^2}\frac{1}{3}\left\langle \nabla^2 \mathrm{U} \right\rangle$$

3. Molecular Systems

Harmonic anisotropic lineshape:

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

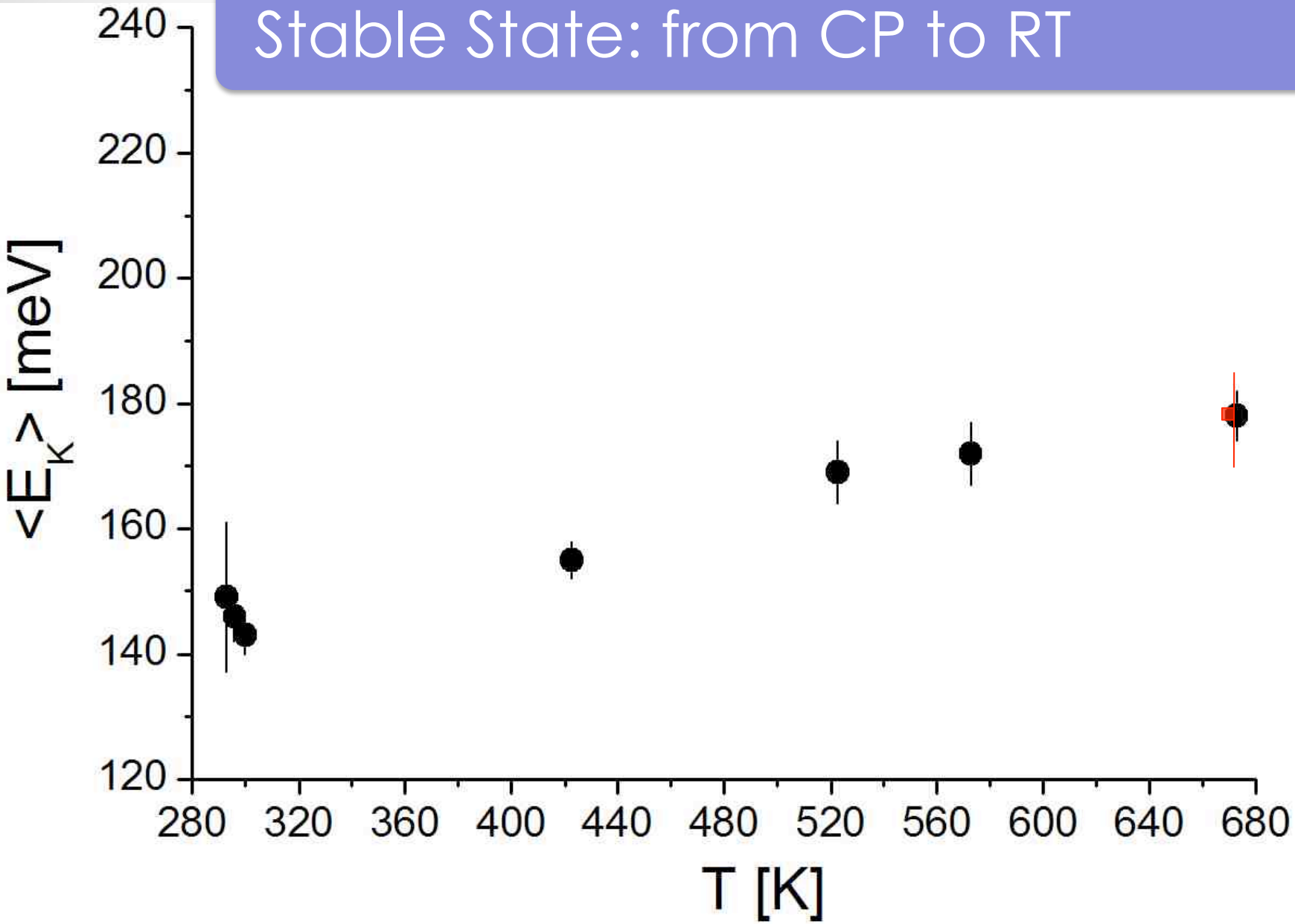
General case:

$$n(p) = \frac{\exp\left(-\frac{p^2}{2\,\sigma^2} \right)}{\left(\sqrt{2\pi\,\sigma^2} \right)^3} \sum_{n \geq 2} \left[1 + (-1)^n a_n L_n^{\frac{1}{2}} \left(\frac{p^2}{2\sigma^2} \right) \right]$$

Examples

…

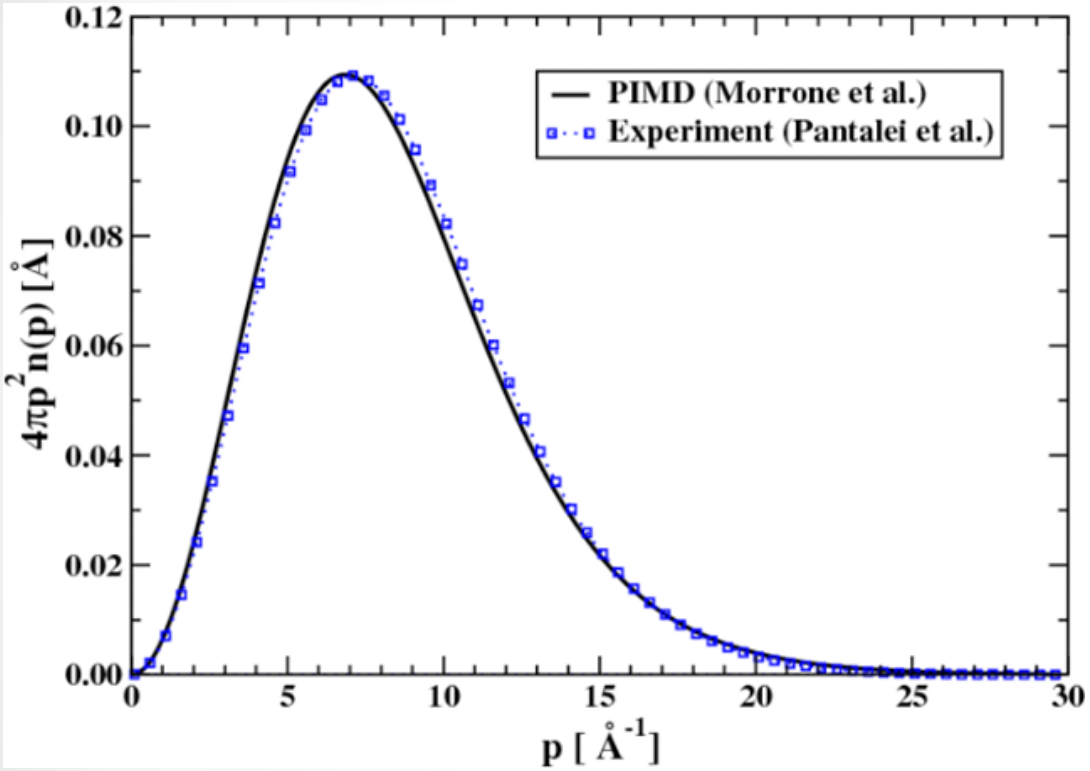
Stable State: from CP to RT



C. Pantalei et al., PRL 100, 177801 (2009)

C. Andreani et al JCP. 115, 11243, (2001).

Supercritical water



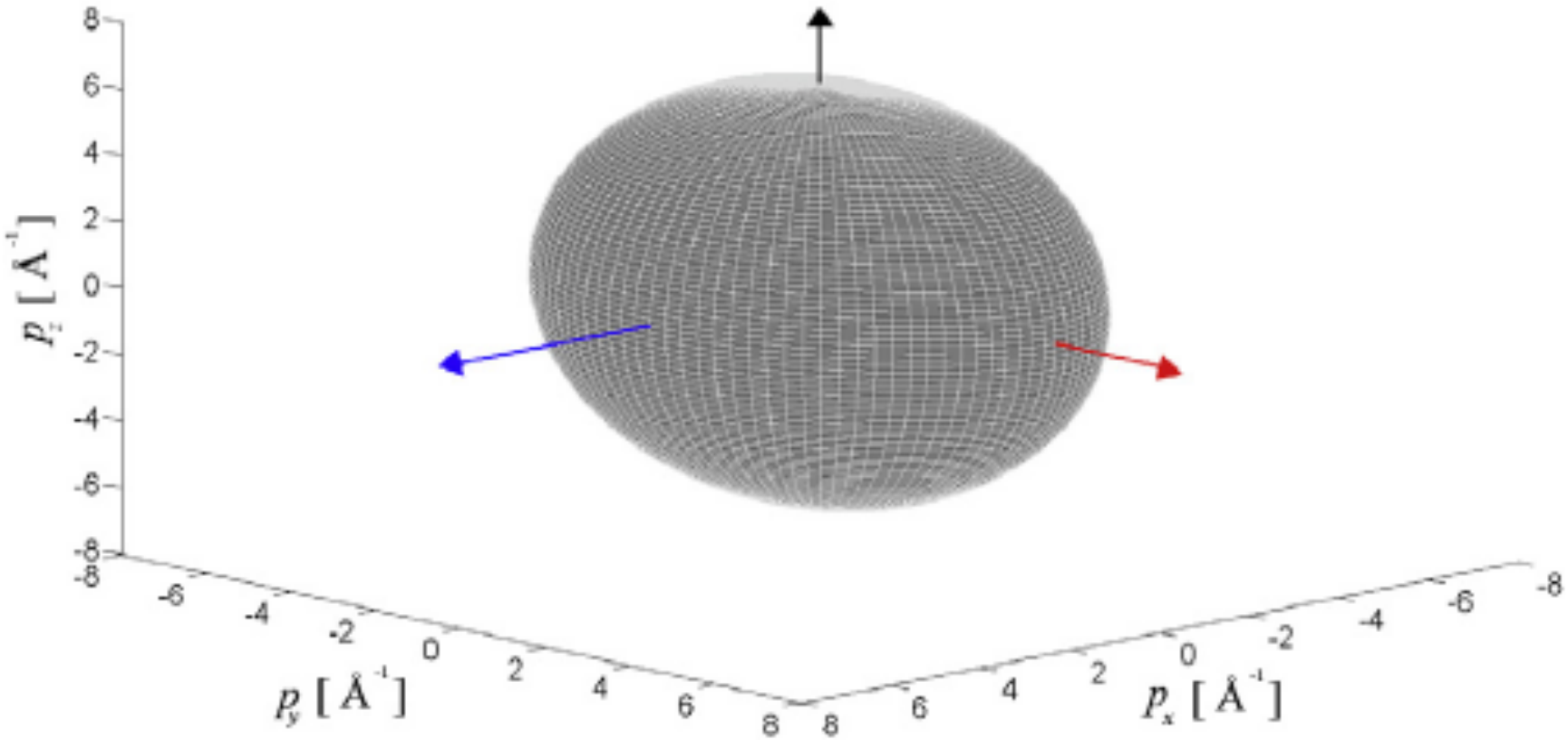
DINS Exp: C. Pantalei et al.
PRL **100** 177801 (2008):
T= 673 K
P=106 MPa
r=0.7 g/cm^3

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

n(p) harmonic- anisotropic lineshape :

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

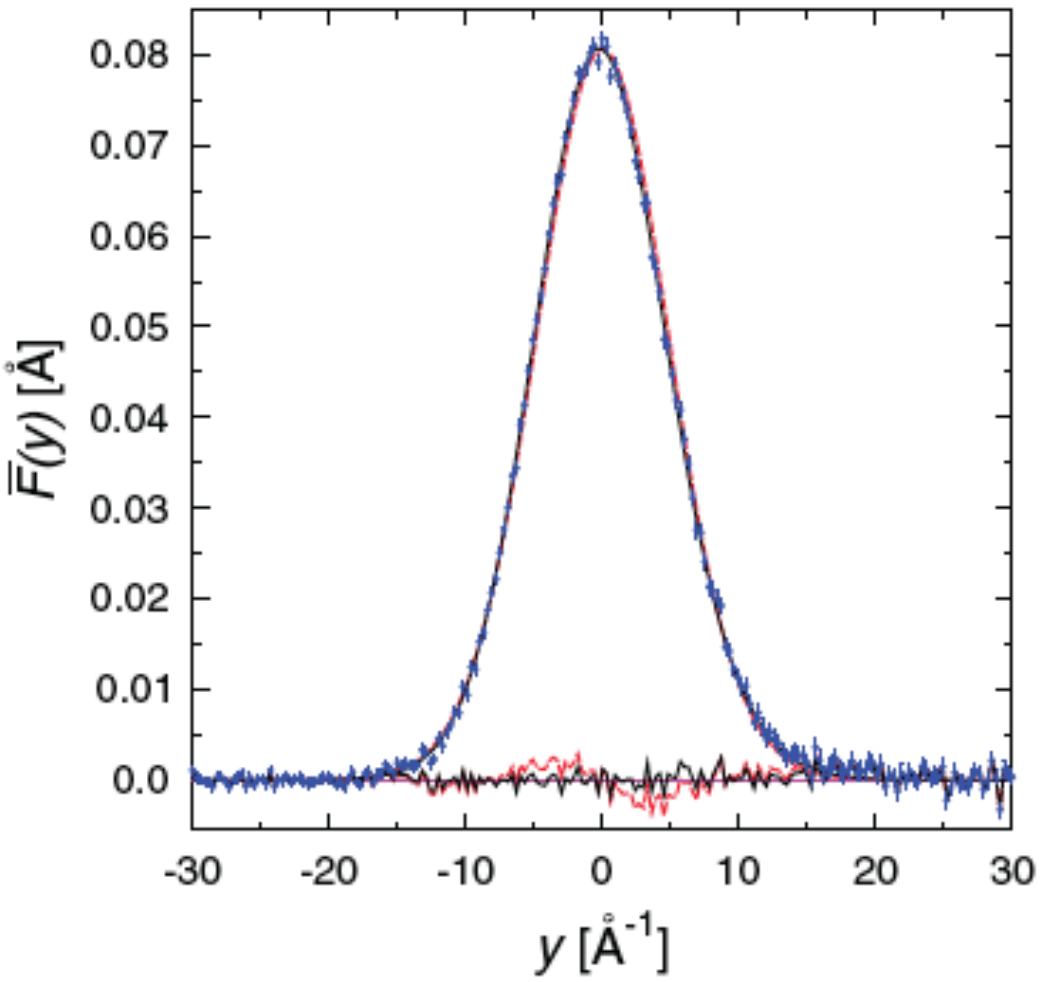
Stable State: water CP



igure 2. Momentum ellipsoid for $n(p)$ in thermodynamically stable water at $T = 673$ K (see main text for details).

H quantum effects in ice

ICE



DINS at T= 271 K

2.5 eV≤ ω ≤ 30 eV

$$F_l(y,q) = [J_{IA}(y) + \Delta J_l(y,q)] \otimes R_l(y,q),$$

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

ICE

DINS at 271 K ●

M1 fit ------

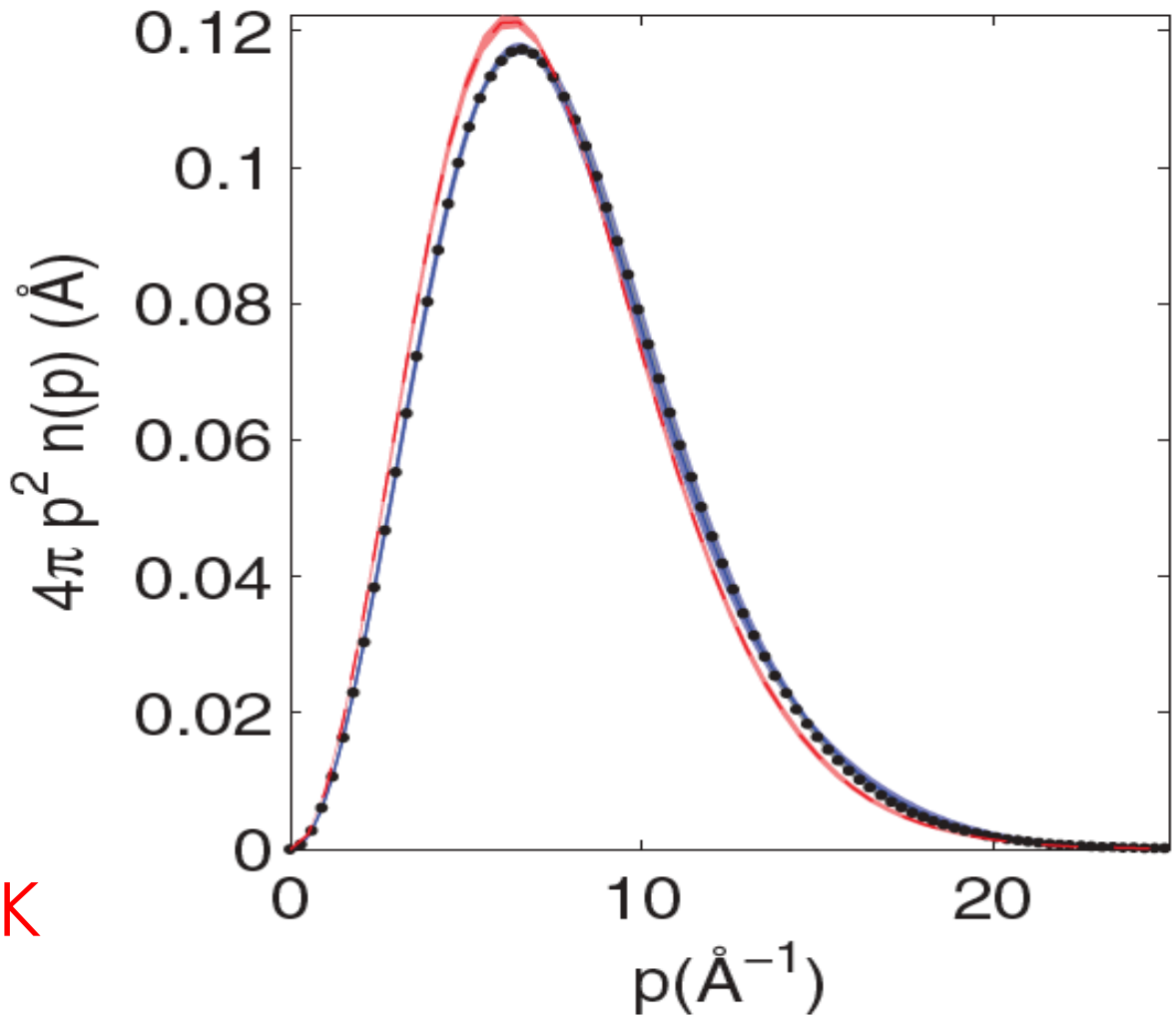
$\langle E_K \rangle$ = 156±2 meV

M2 fit ___

$\langle E_K \rangle$ = 154±2 meV

*PICPMD ---- at 269 K

$\langle E_K \rangle$ = 143 ±2 meV

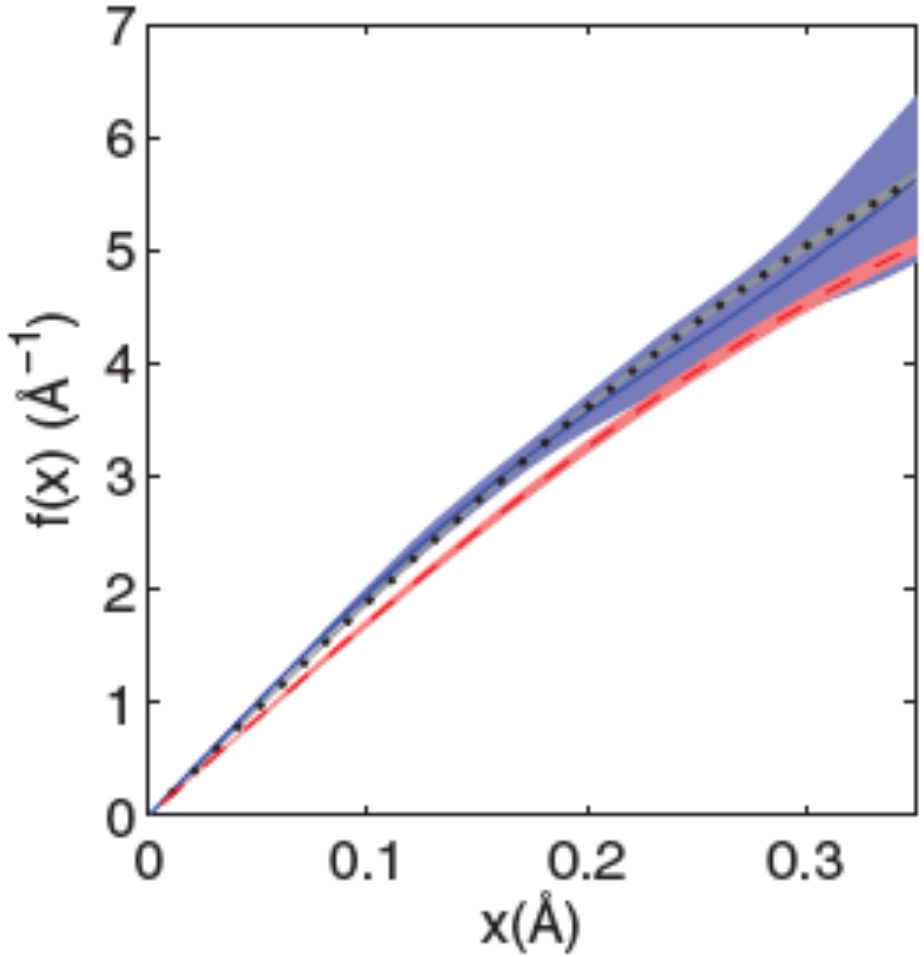


*L. Lin, J. A. Morrone, R. Car and M.Parrinello, *Phys. Rev.* B **83**, 220302(R) (2011).

ICE

$$f(x) = [-\log \ n(x)]' - \frac{Mx}{\beta\hbar^2}$$

$$f(x) = -\frac{Mx}{\beta\hbar^2} + \frac{\int_0^\infty dy \ \ y \ \ \sin(\frac{xy}{\hbar})\bar{J}_{IA}(y)}{\hbar\int_0^\infty dy \ \ y \ \ \cos(\frac{xy}{\hbar})\bar{J}_{IA}(y)}$$



$J_{IA}(y)$ (blue solid line)
M2 (black dots)

***PICPMD (red dashed line)**

*L. Lin, J. A. Morrone, R. Car, and M. Parrinello, *Phys.Rev. Lett.* **105**, 110602 (2010).

Something we did not talk about

Filter Spectrometers for IS at low q and high ω

- ❖ What do we measure
- ❖ Resolution components

Data corrections Filter Spectrometers

- ❖ Correction for Gamma background
- ❖ Correction for multiple scattering

Future development on VESUVIO

Comparison of resolution of chopper and resonance filter spectrometers at eV energies: complementary use of FS and Chopper spectrometers for n(p)

NCS in polyatomic systems, N(P) and n(p)

Reading material

- P.A. Seeger, A.D. Taylor and R. M. Brugger, Nucl. Instr. Methods A 240, 98 (1985)
- J. M. F. Gunn, C. Andreani and J. Mayers, J. of Physics C, 19, L835 (1986)
- C. Andreani, A. Pietropaolo, R. Senesi, G. Gorini, E. Perelli Cippo, M. Tardocchi, N. J. Rhodes, E. M. Schoonveld, Appl. Phys. Lett. 85, 5454 (2004)
- C. Andreani, D. Colognesi, J. Mayers and G. Reiter. Adv Phys 54 377-469(2005)
- G. F. Reiter and P. M. Platzman, Phys. Rev. B 71, 054107 (2005)
- N. I. Gidopoulos, Phys. Rev. B , 71, 054106 (2005)
- J Mayers Phys Rev Lett 98 049601 (2007)
- R. Senesi, A Pietropaolo et al. Phys. Rev. Lett. 98 138102 (2007)
- A Pietropaolo, R Senesi, C Andreani et al. Phys. Rev. Lett. 100 127802 (2008).

- E. Perelli Cippo, G. Gorini, M. Tardocchi, C. Andreani, A. Pietropaolo, R. Senesi, N. Rhodes, E. Schoonveld, Nuc Inst Meth A, 589, 296 (2008).

- J Mayers and M A Adams Calibration of the electron volt spectrometer VESUVIO at ISIS Rutherford-Appleton :Laboratory Technical Report RAL-TR-2009-022.

- C. Windsor, Pulsed Neutron Scattering, (Taylor and Francis, London, 1981)

- J. Mayers, C. Andreani, G. Baciocco, Phys Rev B39 2022 (1989)

Thank you!