

eV Spectrometers

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SYLLABUS

eV spectroscopy and what do we measure

Introduction

DINS technique

$n(p)$, $\langle E_K \rangle$

Where and How do we make the measurements

Introduction to eV spectrometers

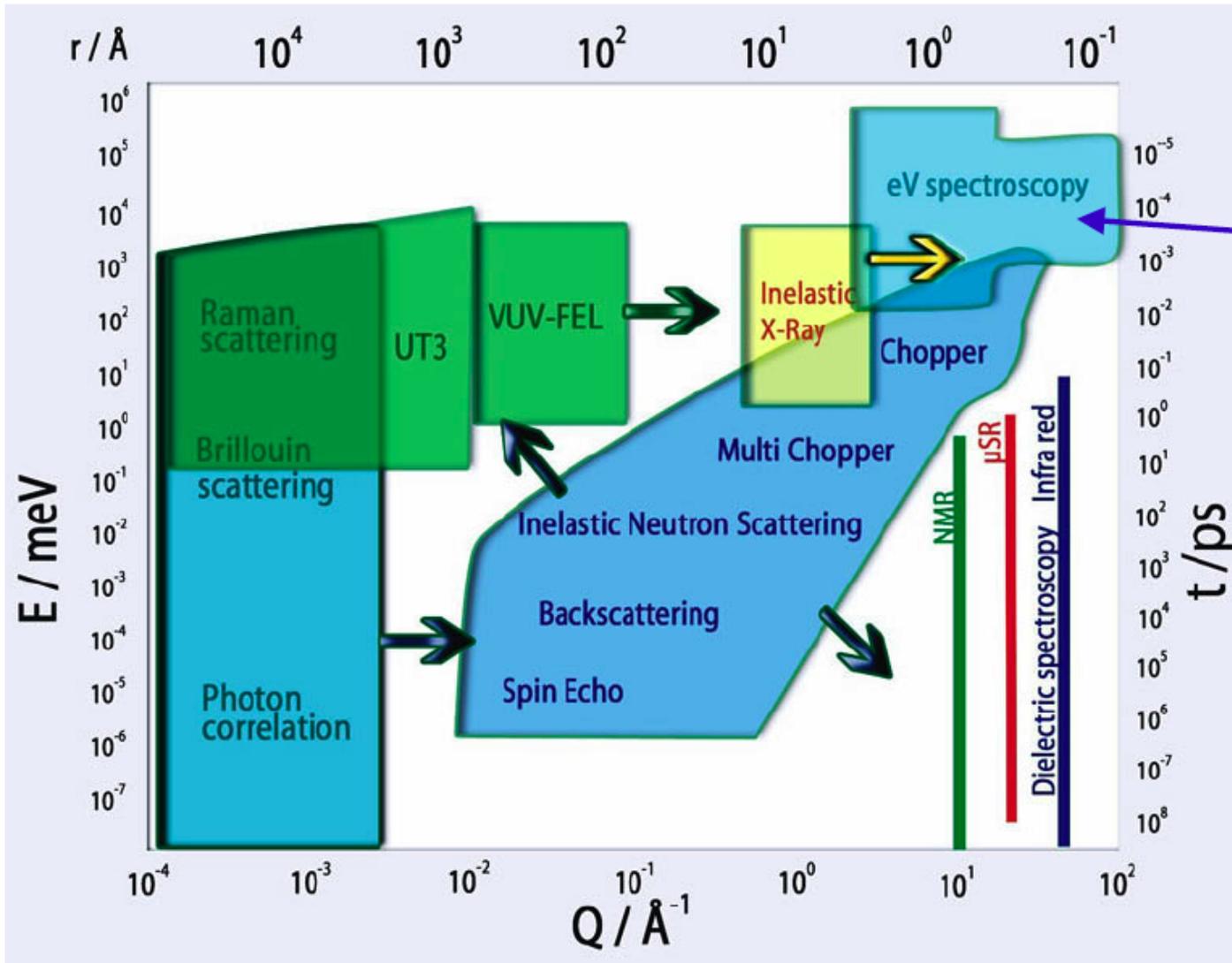
Instrumentation, techniques and methods

What are the obstacles

Examples of scientific results

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

Struck Atoms
travel only a short
time- small distance



implying
large

Q, ω

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

$$\omega > 1 \text{ eV}$$

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

$$0.1 \text{ \AA} < r < 0.2 \text{ \AA}$$

eV SPECTROSCOPY

Deep Inelastic Neutron Scattering

1. Because of Heisenberg indetermination principle **scattered neutron at high q explores regions of the sample of small dimension**. Thus eV neutrons are ideal probe to study single particle properties, (no coherent effects from collective dynamics inside the system) (Incoherent Approximation). The distance over which the neutron phase change appreciably is much lower than the typical interparticle distance d :

$$\frac{2\pi}{q} \ll \bar{d}$$

2. **High ω implies**, because of the time-energy indetermination principle, that the **scattering process occurs in a very short time** (Impulse Approximation, IA).

1. DINS (Deep Inelastic Neutron Scattering)

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 of struck particle

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

V.I. Gol'danskii, Soviet Phys. JETP 4 604 (1957)

P.C. Hohenberg and P.M. Platzmann, Phys. Rev. 152 198 (1966)

2. DINS

Inelastic neutron scattering cross section expressed in terms of $n(\mathbf{p})$ → **Impulse Approximation**

$$S_{\text{IA}}(\mathbf{q}, \omega) = \frac{M}{\hbar q} J(y, \hat{\mathbf{q}})$$

$$\frac{\hbar q}{M} S_{\text{IA}}(\mathbf{q}, \omega) = J(y, \hat{\mathbf{q}}) = \int n(\mathbf{p}) \delta(y - \mathbf{p} \cdot \hat{\mathbf{q}}) d\mathbf{p}$$

**Response Function or
Neutron Compton Profile**



y is the West scaling variable:

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



recoil energy

G.B. West, Phys. Rev. C 18 263 (1975).

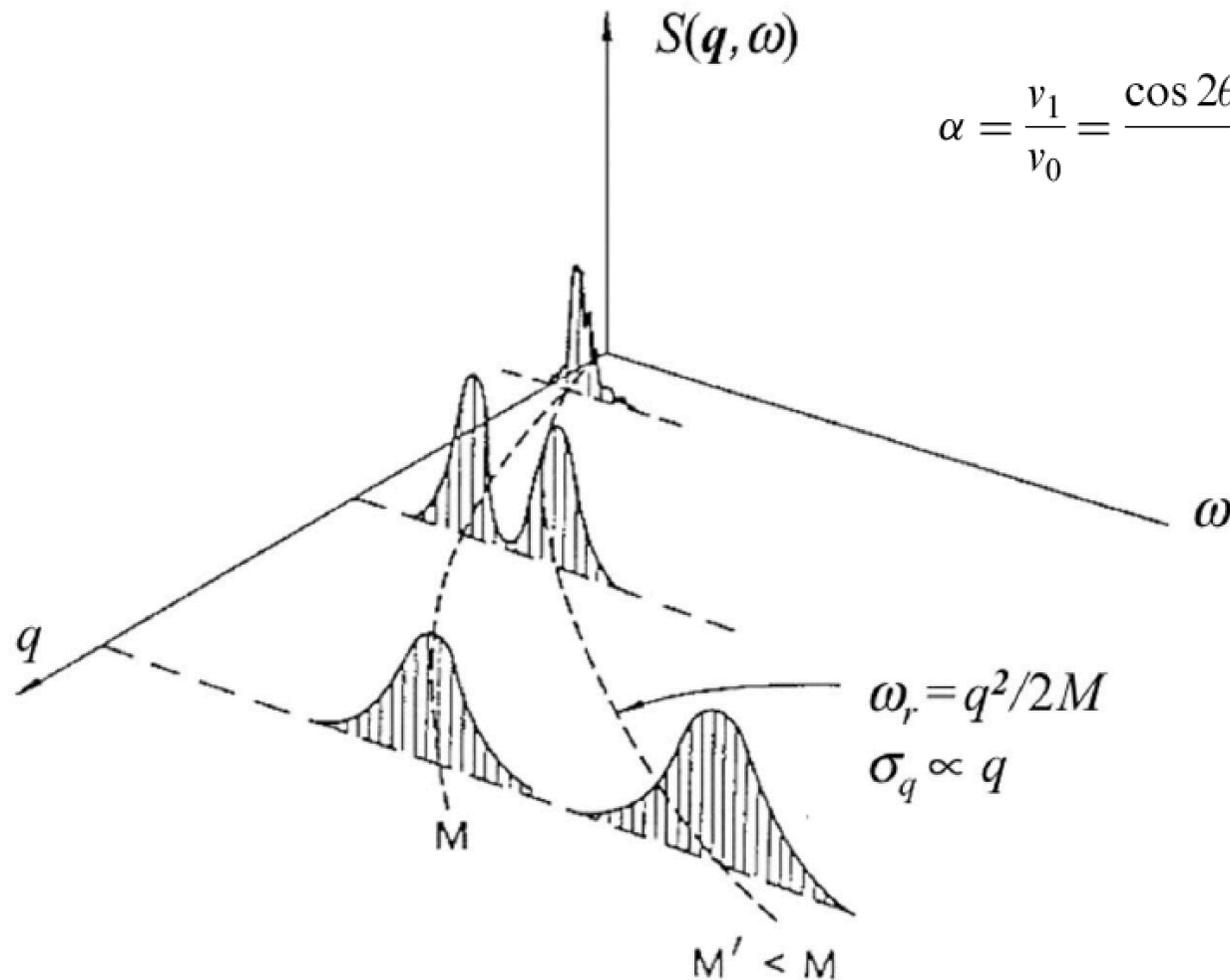
J. M. F. Gunn, C. Andreani, J. Mayers, *J. P. C. Solid State Physics* 19, L835 (1986)

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

$$\alpha = \frac{v_1}{v_0} = \frac{\cos 2\theta + [(M/m)^2 - \sin^2 2\theta]^{1/2}}{(M/m) + 1}$$

$$M = m,$$

$$\alpha = \cos 2\theta/2,$$



3. DINS

At finite q - Final State Effects (FSE)

Interactions among recoiling particle and the surroundings \rightarrow inter- and intramolecular interactions:

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

4. DINS

$$n(\vec{p}) = \left| \int \psi(\vec{r}) \exp(i\vec{p} \cdot \vec{r}) d\vec{r} \right|^2 \quad \langle E_k \rangle = \frac{\langle p^2 \rangle}{2M} = \frac{1}{2M} \int n(\mathbf{p}) p^2 d\mathbf{p}$$

Nuclear Quantum Effects

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

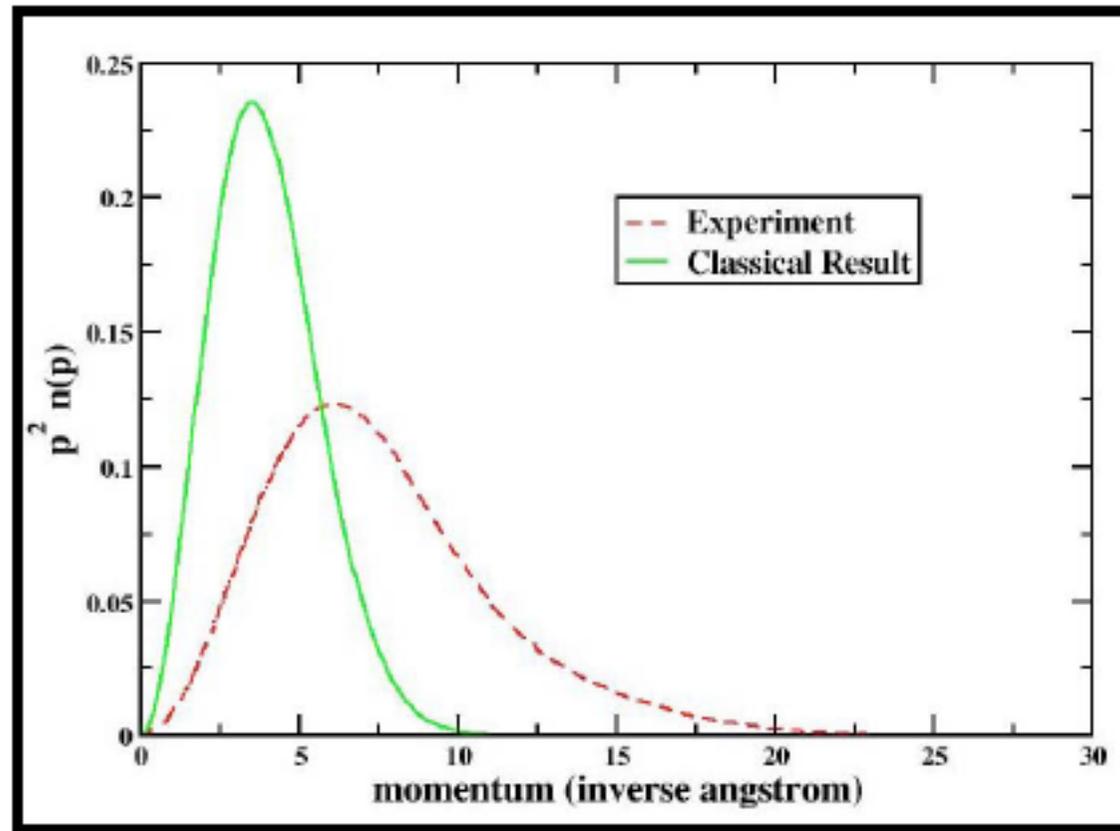


$$n(p) \text{ \& } \langle E_K \rangle \rightarrow \text{PES}$$

$$\left\{ \text{localization} \right\} \rightarrow \text{excess of } \langle E_K \rangle \frac{\langle p^2 \rangle}{2M} > > \frac{3}{2} k_B T$$

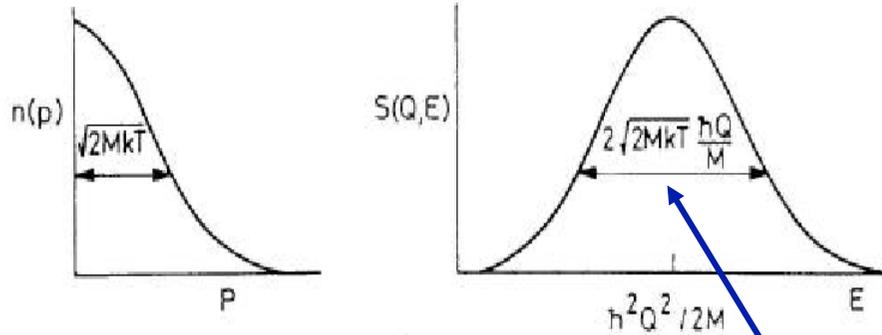
Nuclear quantum effects (NQE's)

Not only the electrons but also the protons need quantum mechanical description. NQE's essential to explain material properties.

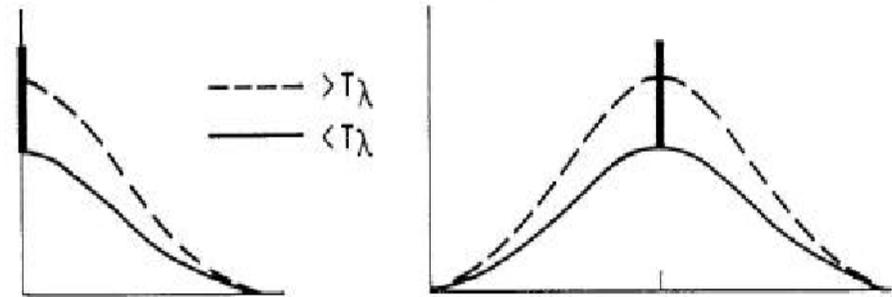


The Proton Momentum Distribution $n(p)$ in water probed by Neutron Compton Scattering (NCS) displays importance of nuclear quantum effects

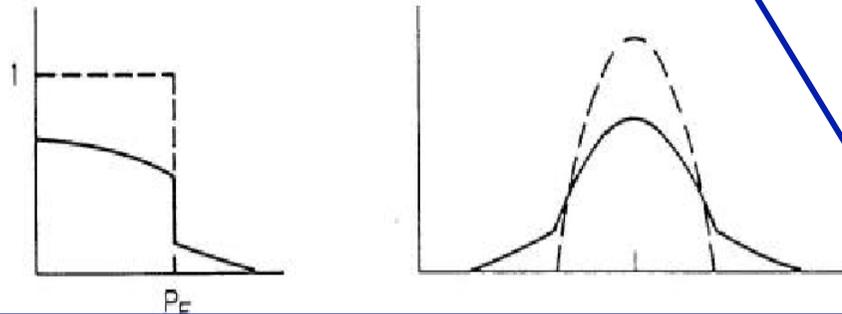
CLASSICAL IDEAL GAS



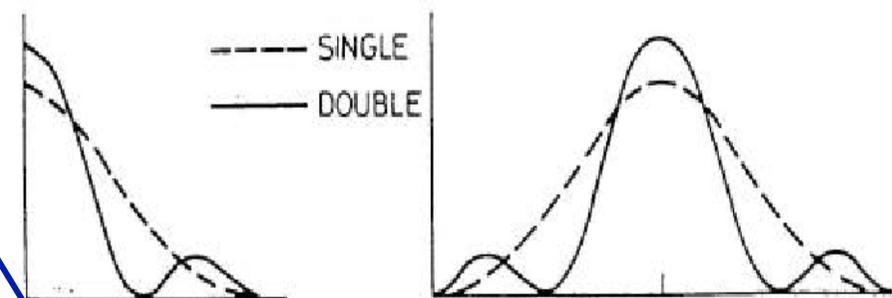
BOSE CONDENSATE (⁴He)



FERMI LIQUID (³He)



PARTICLE IN POTENTIAL WELL



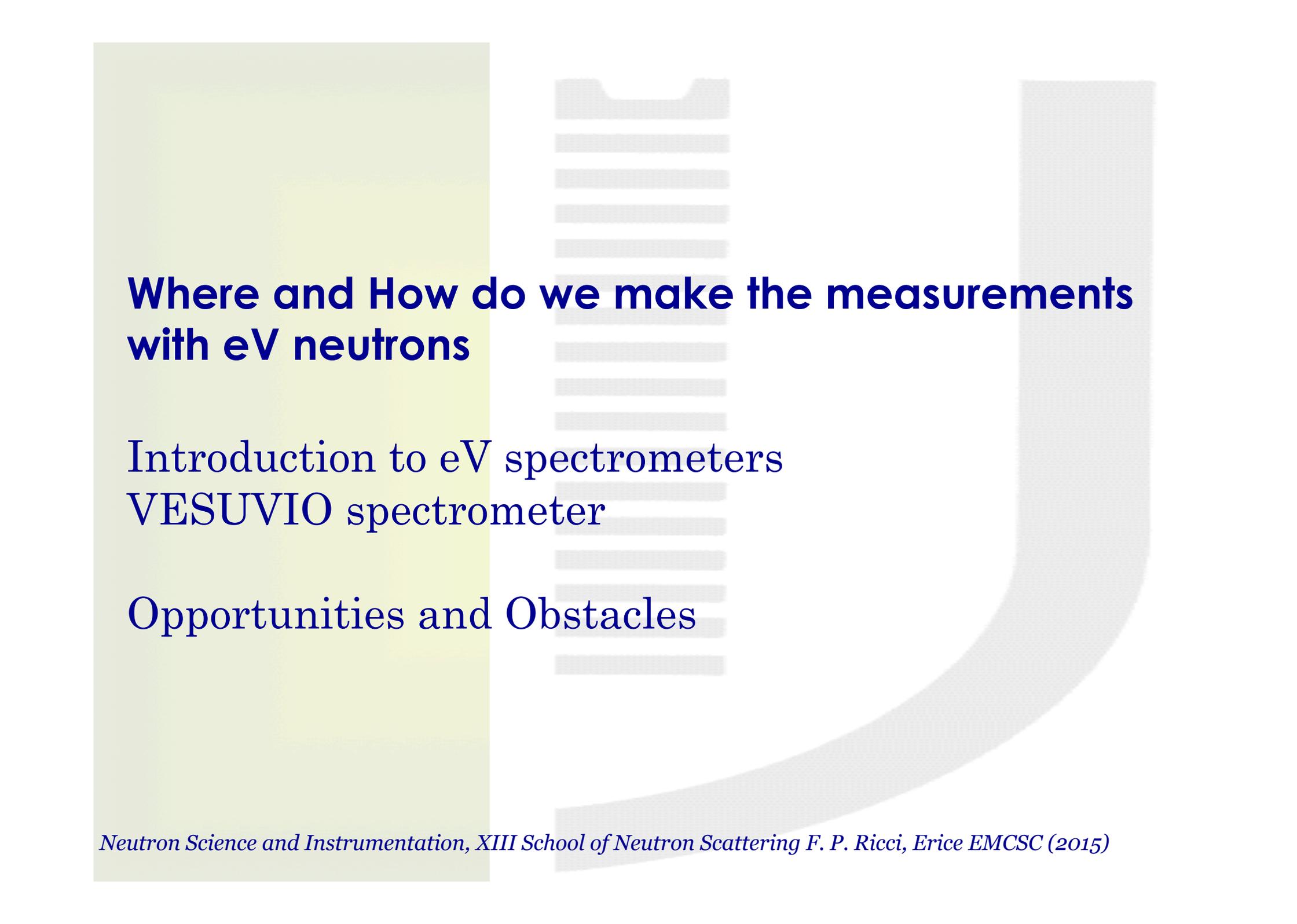
$$n(p) \propto \exp\left[-p^2 / (4Mk_B T)\right] \Rightarrow \sigma^2 = 2Mk_B T \quad \text{Classical system}$$

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

$$\langle E_k \rangle = \frac{1}{2M} \int n(p) p^2 dp$$

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

Momentum Distribution is "Diffraction Pattern" of Wave function



Where and How do we make the measurements with eV neutrons

Introduction to eV spectrometers
VESUVIO spectrometer

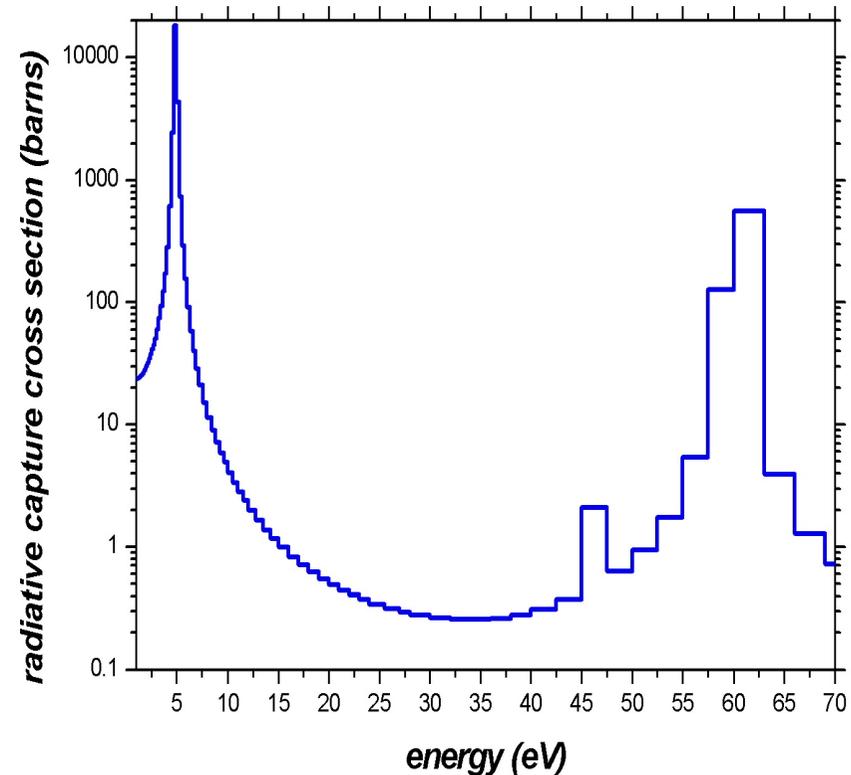
Opportunities and Obstacles

How to select the final neutron energy in the 1-250 eV range

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

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

radiative capture cross section of ^{197}Au



$$\sigma(E) = \frac{\sigma_0}{1 + 4(E - E_R)^2/\Gamma^2}$$

eV Spectroscopy with Direct and Indirect Geometry Spectrometers

FILTER SPECTROMETERS

Resonance Detector Spectrometer (RDS) and Resonance Filter Spectrometer (RFS)

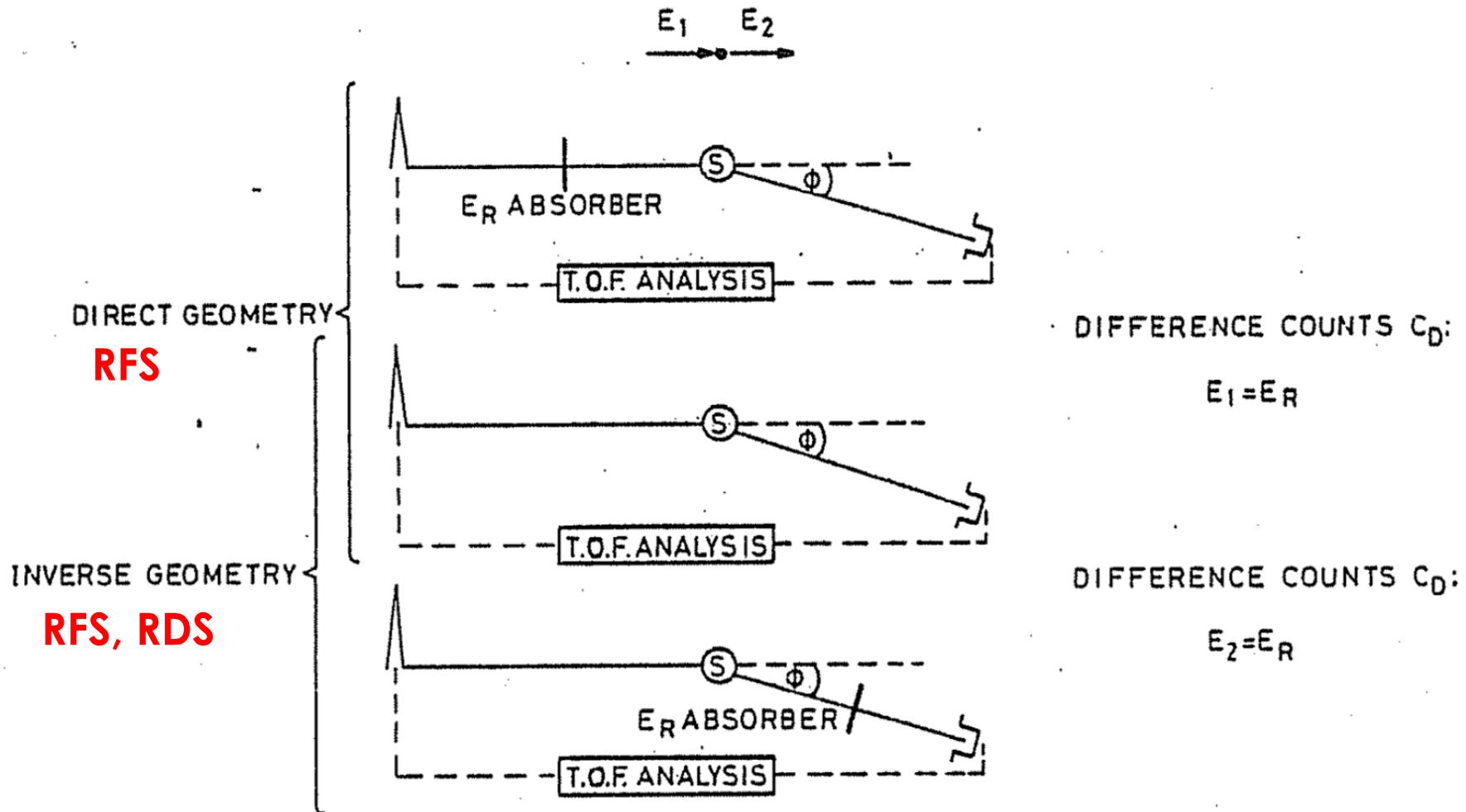


Fig. 1. Principle of resonance filter difference method in direct and inverse geometries.

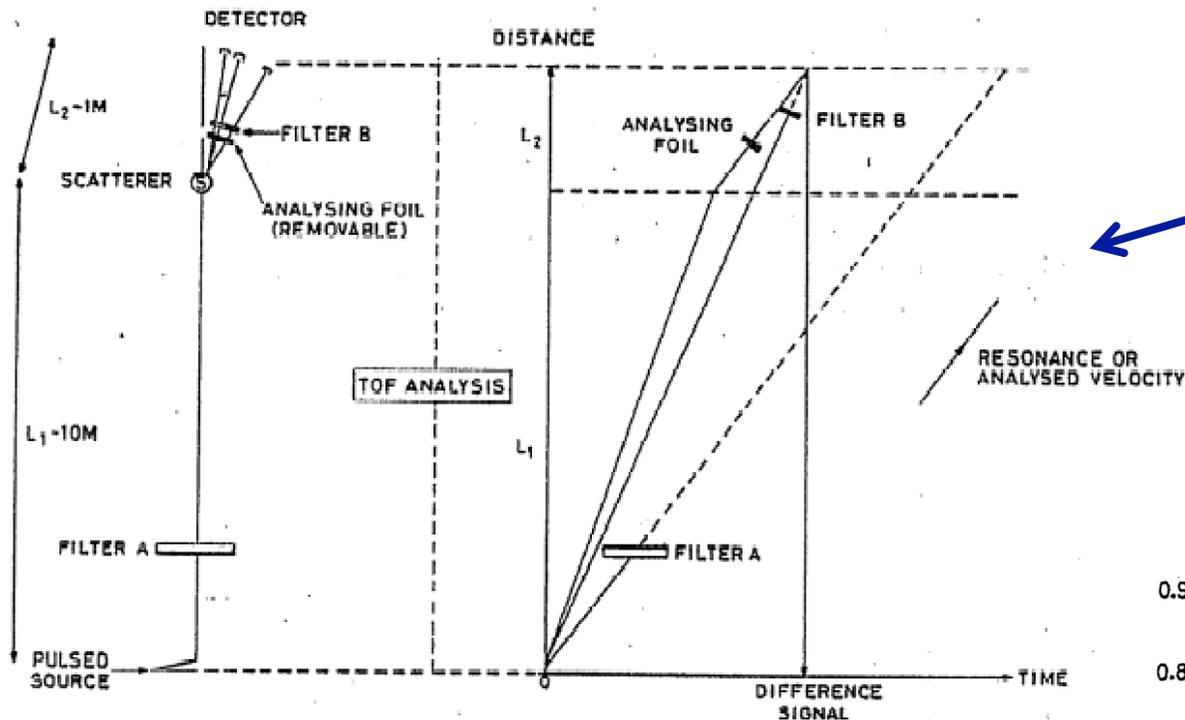
Resonance Filter Spectrometer (RFS)

- ❖ R. M. Brugger, A. D. Taylor, C. E. Olsen, J. A. Goldstone and A. K. Soper, *Proc. 6th Int. Collaboration on Advanced Neutron Source (ICANS-VI)*, Argonne National Laboratory (1982)
- ❖ R. J. Newport, J. Penfold, W. G. Williams, “*Electron Volt Spectroscopy on a Pulsed Neutron Source*”, *Nuclear Instrument and Methods* **224**, 120 (1984)
- ❖ P.A. Seeger, A.D. Taylor and R. M. Brugger, “The Filter Difference Method” *Nucl. Instr. Methods A* **240**, 98 (1985).

Resonance Detector Spectrometer (RDS)

- ❖ R.N. Sinclair, M.C. Moxon and J. M. Carpenter, *Bull. Am. Phys. Soc.* **22** 101 (1977)
 - ❖ D. R. Allen, E. W. J. Mitchell and R. N. Sinclair, *J. Phys.* **E13**, 639 (1980)
 - ❖ J. M. Carpenter, N. Watanabe, S. Ikeda, Y Masuda and S. Sato, *Nuclear Instrument Methods*, **120**, 126 (1983).
 - ❖ J. M. Carpenter and N. Watanabe, *Proc. of the 1984 Workshop on high Energy Excitations in Condensed Matter*, Los Alamos (1984)
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Principles of RFS

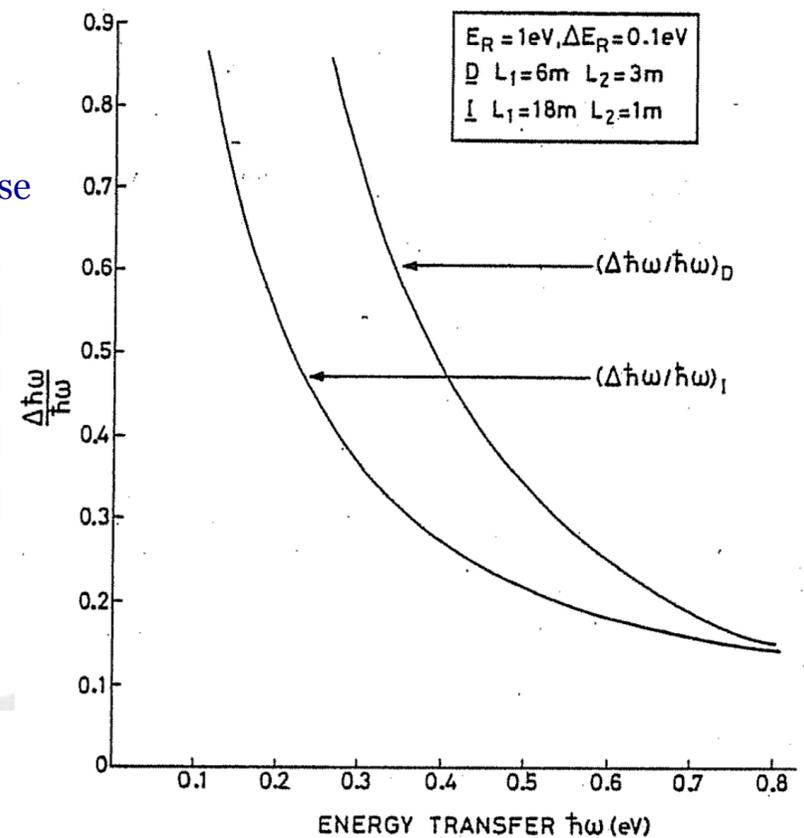


RFS: Schematic diagram and the principle of the method in Inverse Geometry

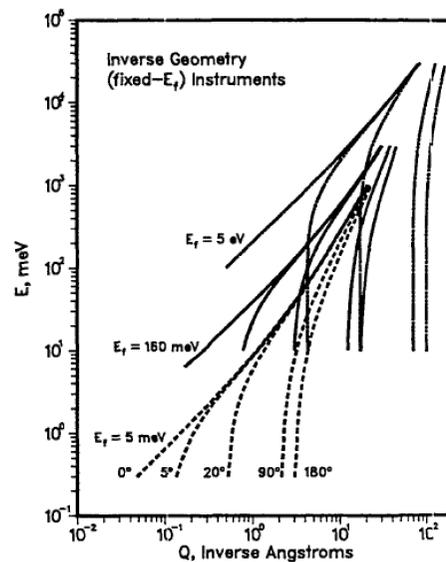
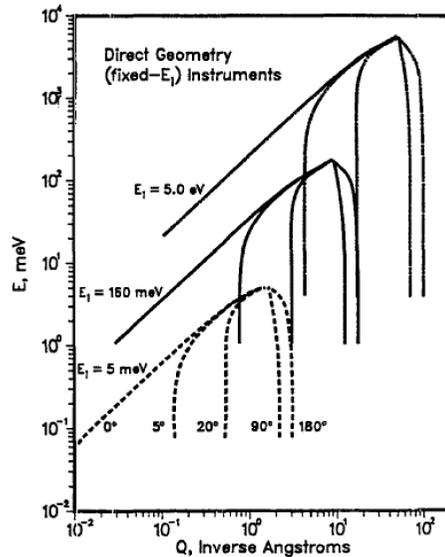
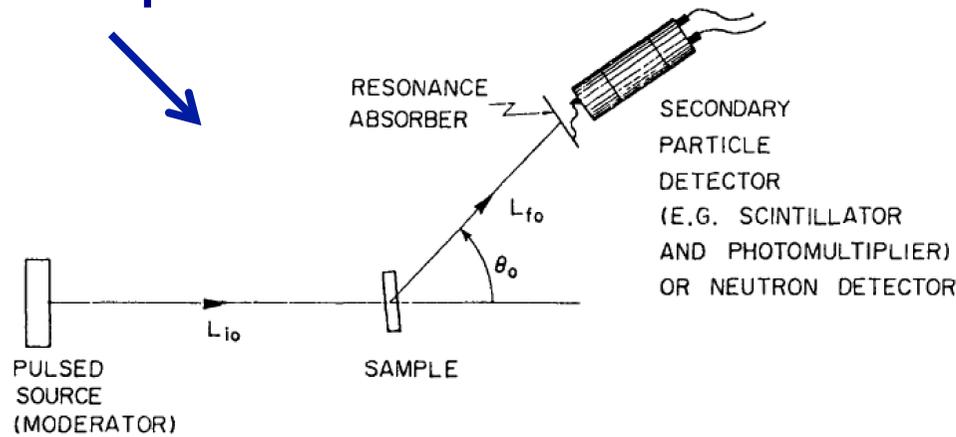
Table 1
Potentially useful resonances for filter difference method

Isotope	Natural abundance	E_R (eV)	I' (meV)	Δ_{D293} (meV)
^{149}Sm	0.138	0.872	61	25
^{240}Pu	-	1.056	33	21
^{242}Pu	-	2.67	27	34
^{181}Ta	1.0	4.28	57	50
^{121}Sb	0.573	6.24	90	73
^{238}U	0.993	6.67	28	54

“Electron Volt Spectroscopy on a Pulsed Neutron Source”
R. J. Newport, J. Penfold, W. G. Williams, *Nuclear Instrument and Methods* **224** 120 (1984).



Principles of RDS



A2 Range of Q , ϵ space covered by various types of spectrometers in energy-loss spectroscopy. Part a, fixed E_i ; part b, fixed E_f spectrometers.

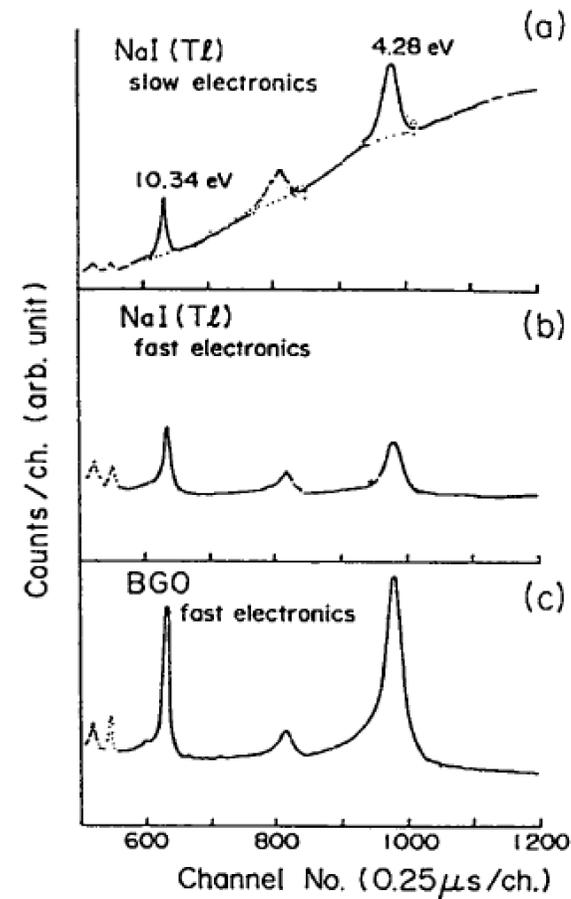
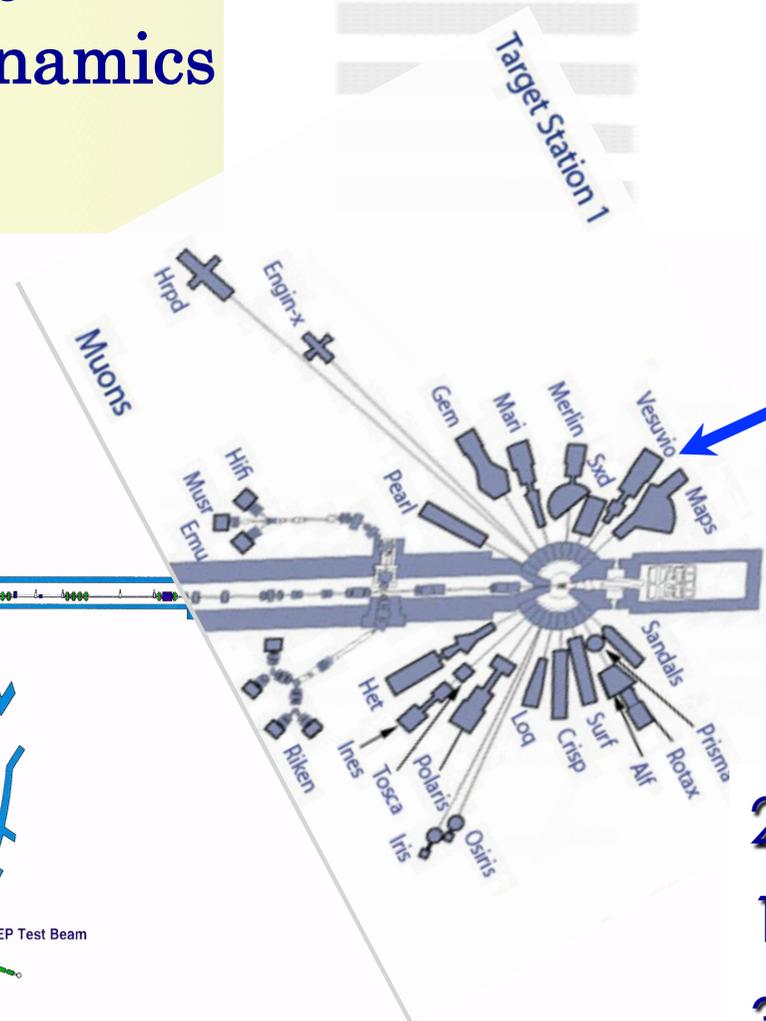
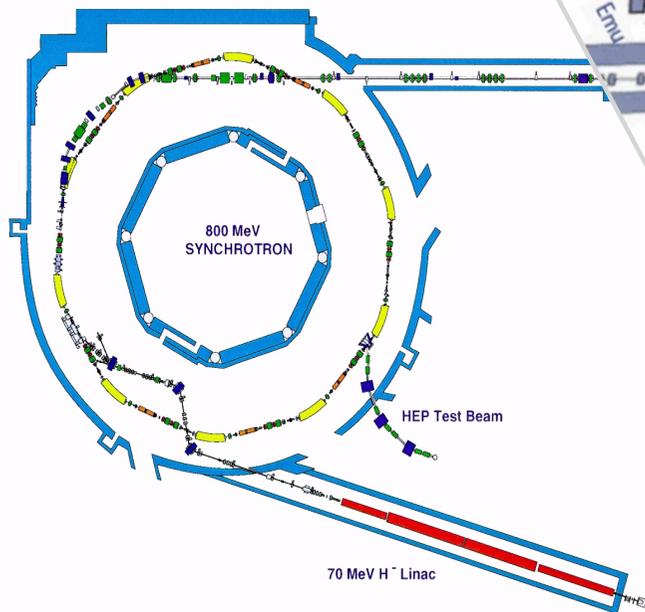


Fig. 2 Preliminary results of the time spectra of the gamma-ray counts, measured by a NaI(Tl) with a slow electronics (a), by the NaI(Tl) with a fast electronics (b), by a BGO with the fast electronics (c). Scattering sample is Pb and the resonance foil is Ta.

J. M. Carpenter and N. Watanabe, *Proc. of the 1984 Workshop on high Energy Excitations in Condensed Matter*, Los Alamos (1984)

5. VESUVIO at ISIS

Single particle
short-time dynamics



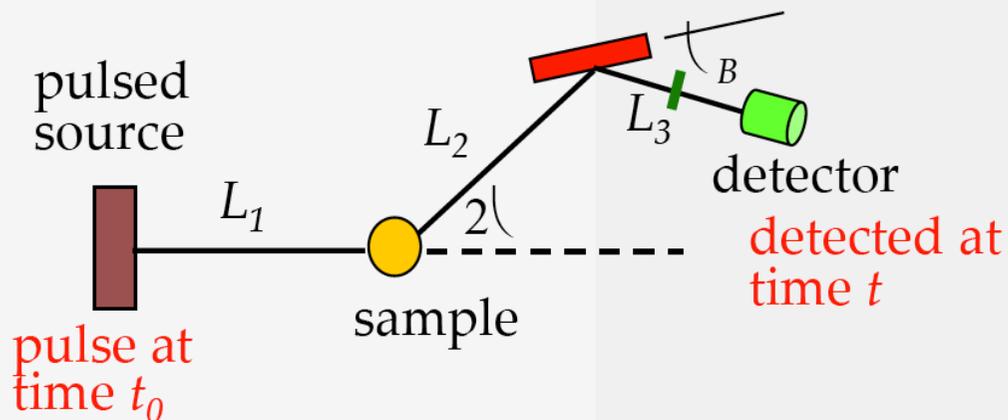
Vesuvio is here

$$20 \text{ \AA}^{-1} < q < 250 \text{ \AA}^{-1}$$
$$1 \text{ eV} < \omega < 200 \text{ eV}$$
$$30^\circ < 2\theta < 160^\circ$$

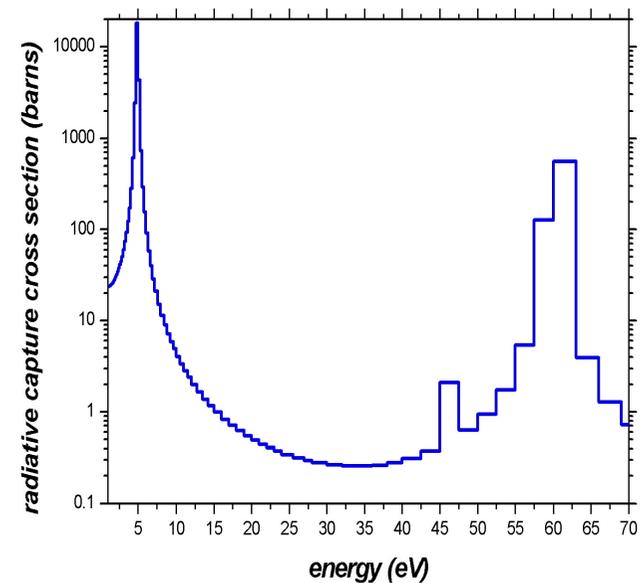
VESUVIO is a filter spectrometer (at eV energy)

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

TOF – xtal (inverse-geometry)



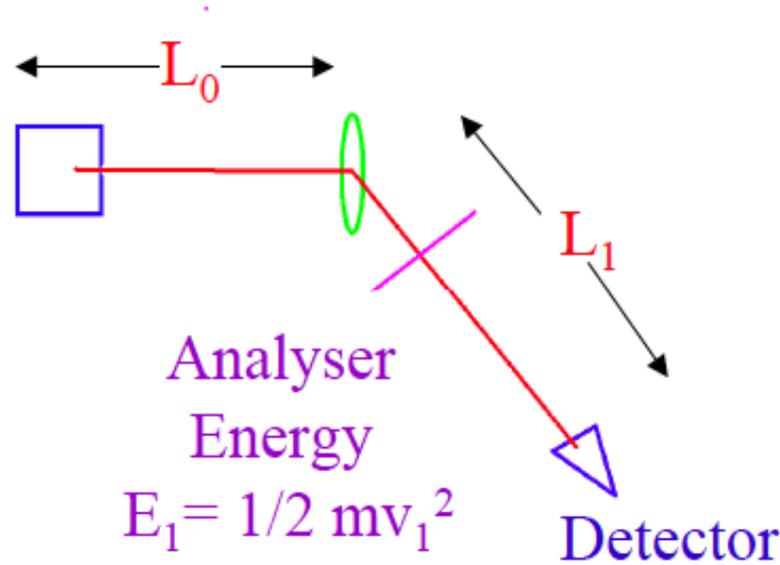
radiative capture cross section of ^{197}Au



attering F. P. Ricci, Erice EMCSC (2015)

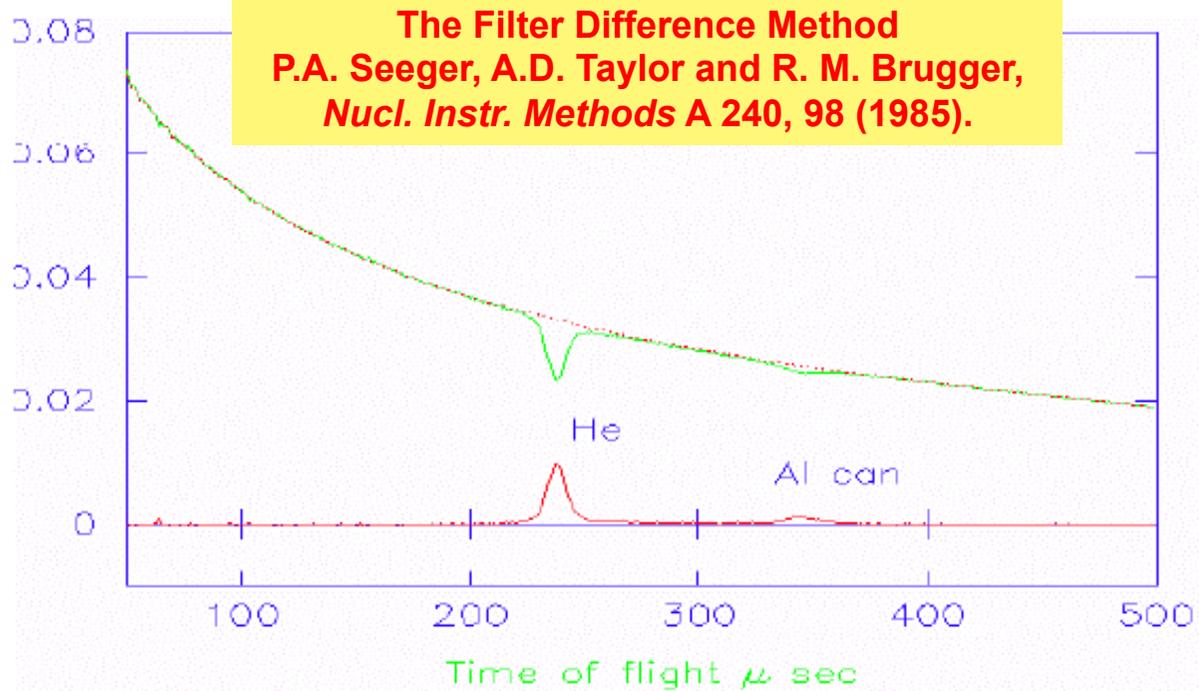
RFS

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

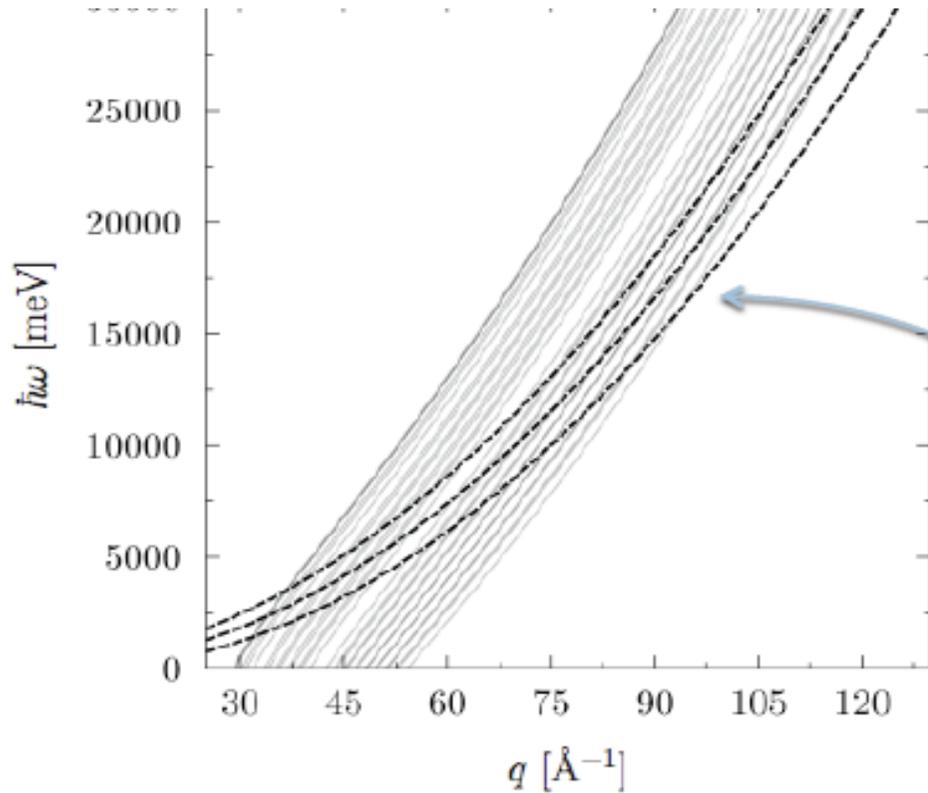


$$\hbar\omega = \frac{1}{2} m (v_0^2 - v_1^2)$$

$$q^2 = m^2 (v_0^2 + v_1^2 - 2v_0v_1 \cos\theta)$$



Incident neutrons of all available energies are generated at $t=0$
Scattered neutrons are energy selected by the filters and recorded by neutron detector



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

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

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

Count rate as a function of t

CMCSN

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

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

probability that a neutron of energy E_1 is detected

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

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

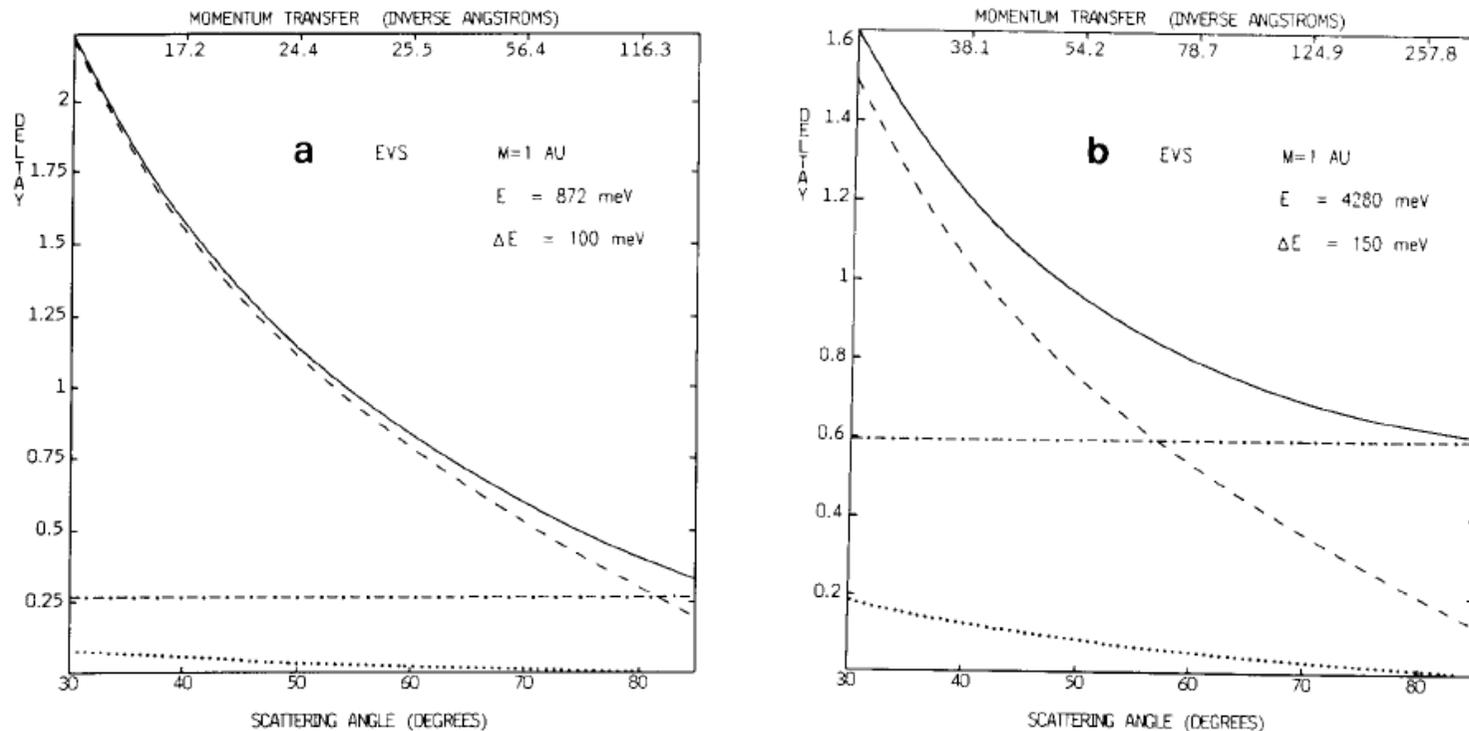


Fig. 2. The total resolution (—) and the energy (---), angular (-·-·-) and timing (·····) contributions to the resolution in hydrogen ($M=1$ amu) shown as a function of scattering angle and momentum transfer for eVS at (a) $E_1 = 872$ meV and (b) $E_1 = 4280$ meV.

RESOLUTION COMPONENTS

Geometrical \mapsto Gaussian

Energy \mapsto Gaussian&Lorentzian

- **single difference (SD)**

example U foil

U resonances: \mapsto 6.7eV, 20.7eV, 37eV

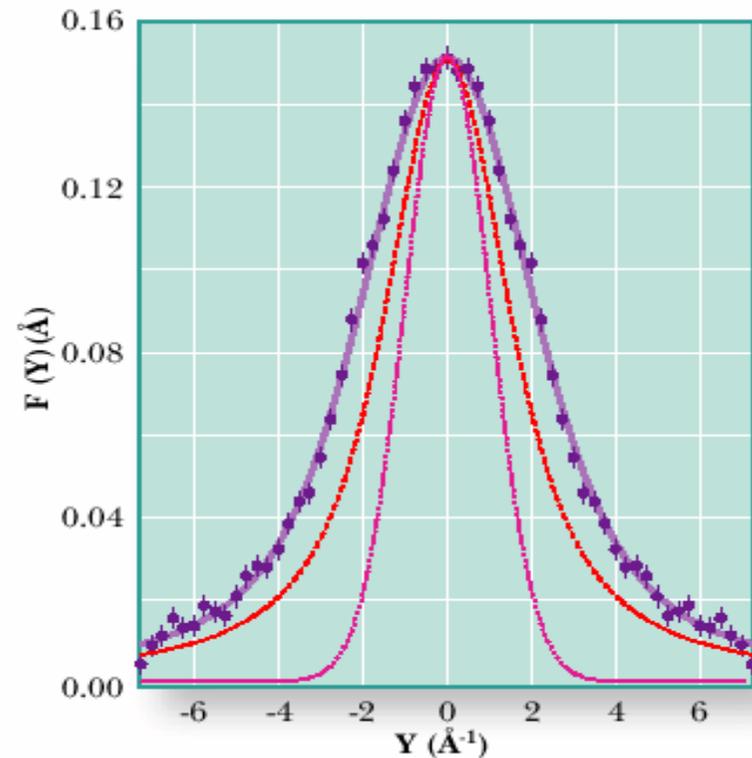
FWHM (at 6.7 eV) \mapsto 0.04 eV

Doppler broadening

at RT \mapsto 0.11 eV

at 70 K \mapsto 0.06 eV

$$\sigma(E) = \frac{\sigma_0}{1 + 4(E - E_R)^2/\Gamma^2}$$



Scattering function $F(y)$ for the ^3He bcc solid sample. Data (full circles); best fit (purple line); resolution function SD (red line); resolution function DD (light purple line)

From Pb sample: VESUVIO resolution determined by fitting Lorentzian \otimes Gaussian convolution to the data and subtracting the Gaussian component, due to intrinsic width of the Pb sample.

DOUBLE DIFFERENCE TECHNIQUE

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

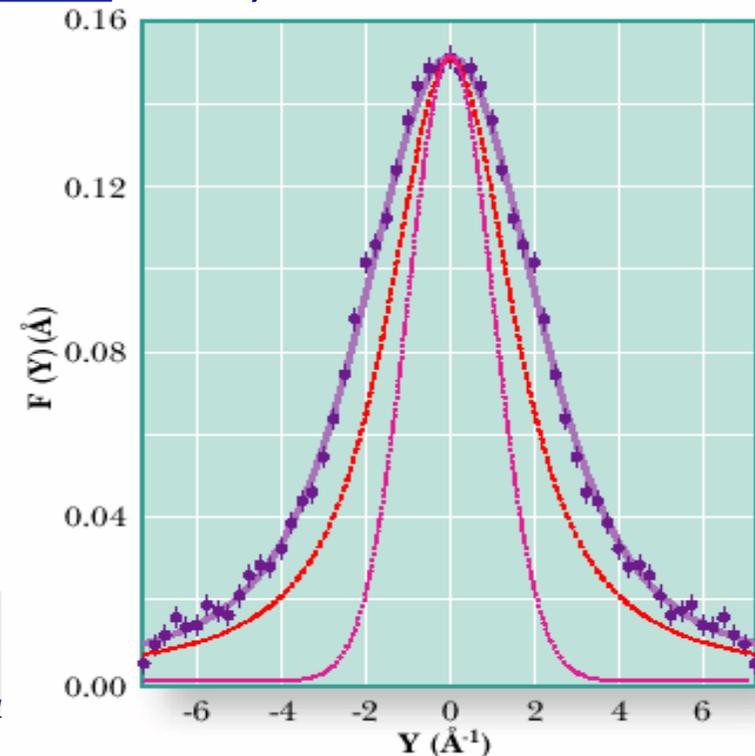
$$1 - T_1(E) = 1 - \exp[-Nt_1\sigma(E)] \sim Nt_1\sigma(E)$$

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

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

★ Resolution reduction of
 ~ 2 for U and Au foils

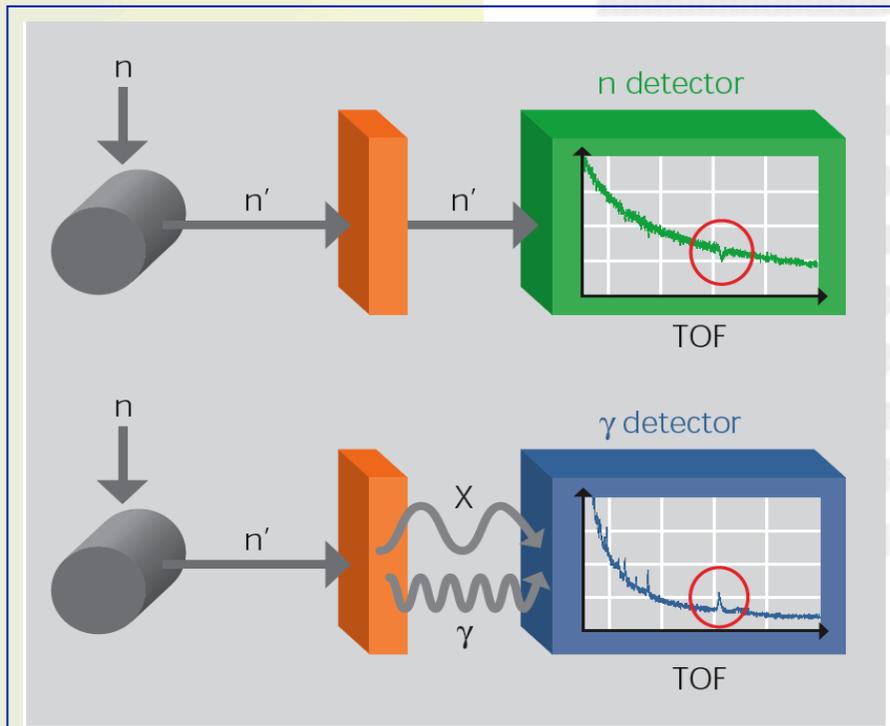
Scattering function $F(y)$ for the ^3He bcc solid sample. Data (full circles); best fit (purple line); resolution function SD (red line); resolution function DD for VESUVIO (light purple line).



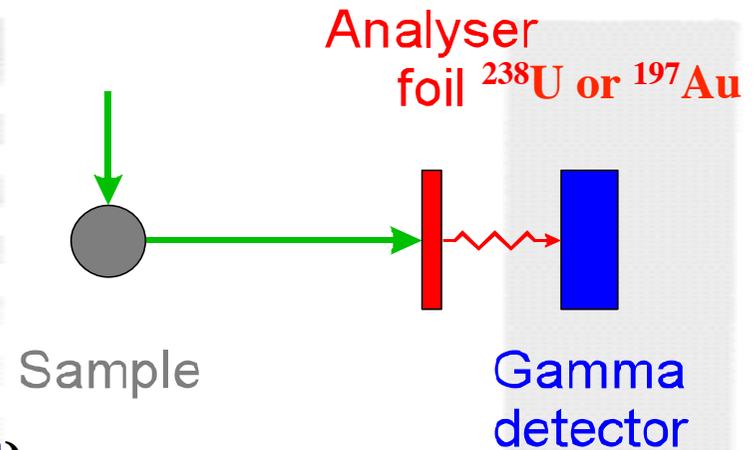
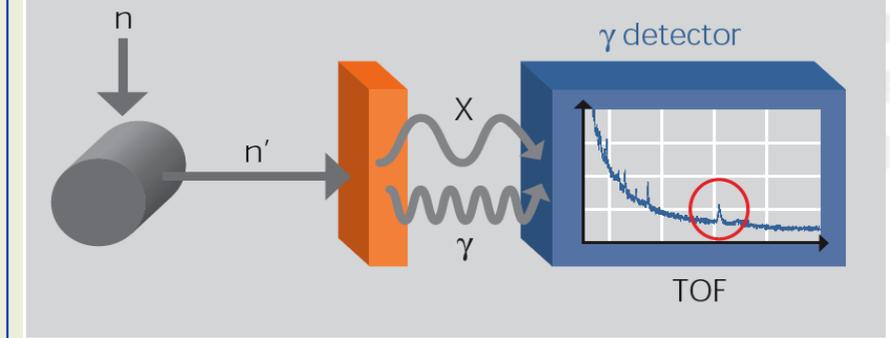
RFS & RDS (from 2002) ON VESUVIO

Principles of:
Resonance Filter Spectrometer (RFS)
Resonance Detector Spectrometer (RDS)

RFS



RDS



**⁶Li-glass
Neutron Detectors**

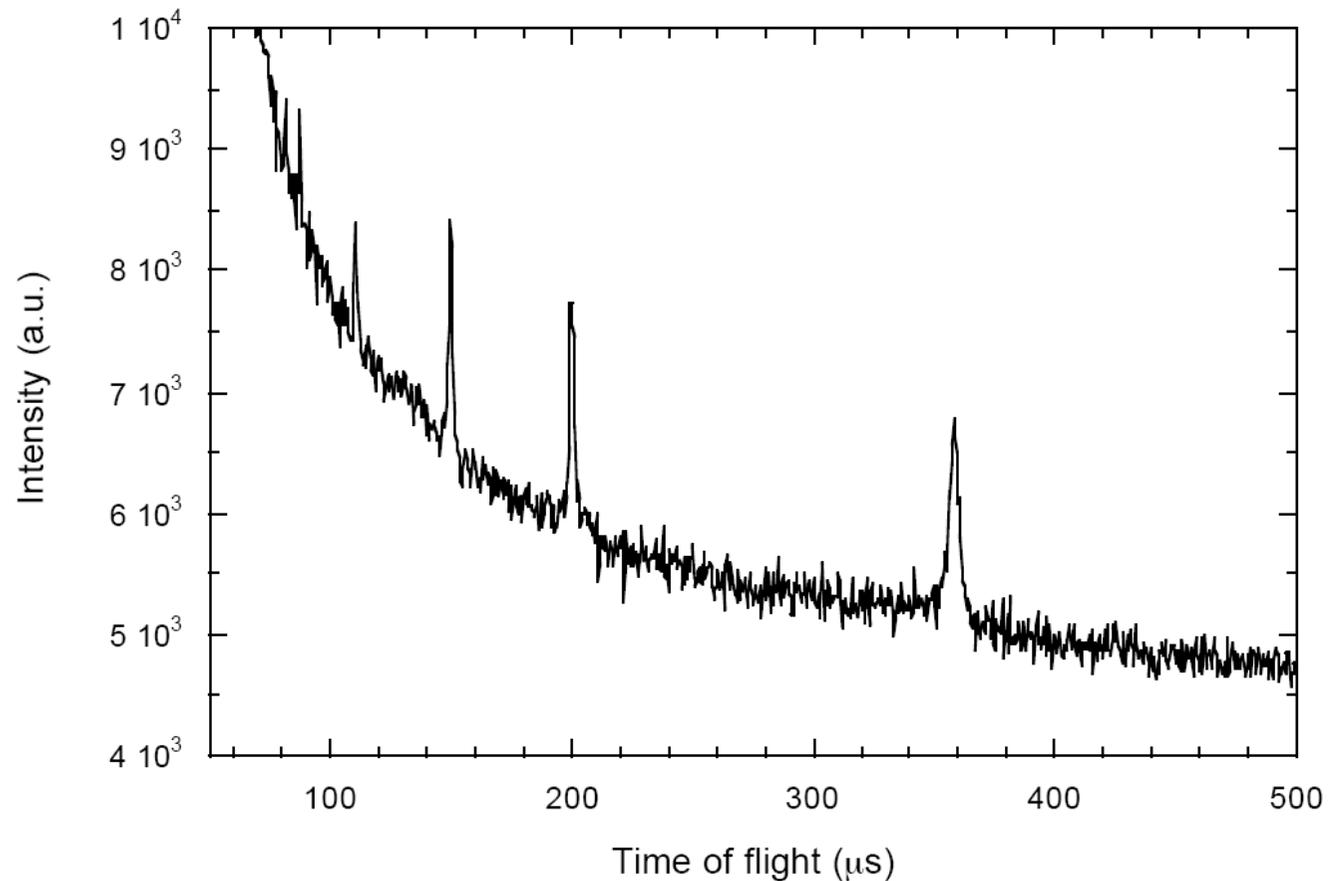


YAP (YAIO₃) scintillator => both the time of flight and the energy E1 of the absorbed neutron are determined when a γ ray is detected

The Resonance Detector Spectrometer, 2002

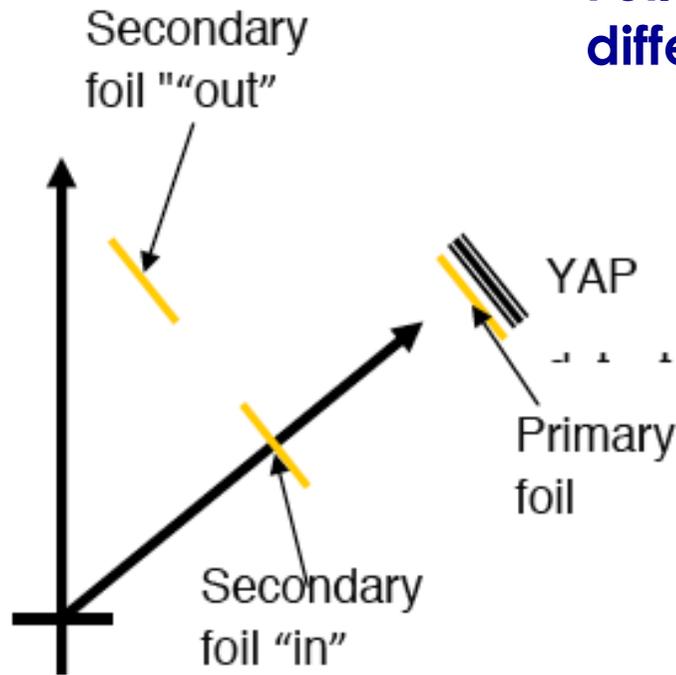
- YAP detector at 2°

Ice at 270K, ^{238}U



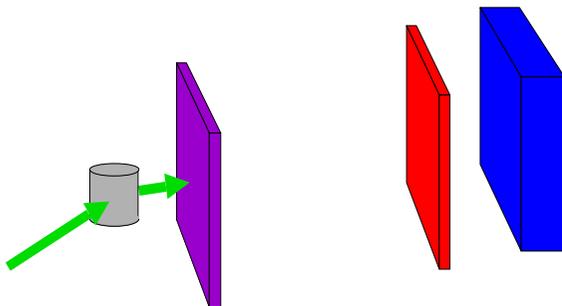
M. Tardocchi et al Nuclear Inst. Methods A 526, 477–492 (2004); Appl. Phys. A: Materials Science & Processing 74, [Suppl.], S189–S190 (2002); G. Gorini et al Nuclear Instruments and Methods A 529, 293-300 (2004)

Foil Cycling Technique: "raw" data is the difference between foil out and foil in data.

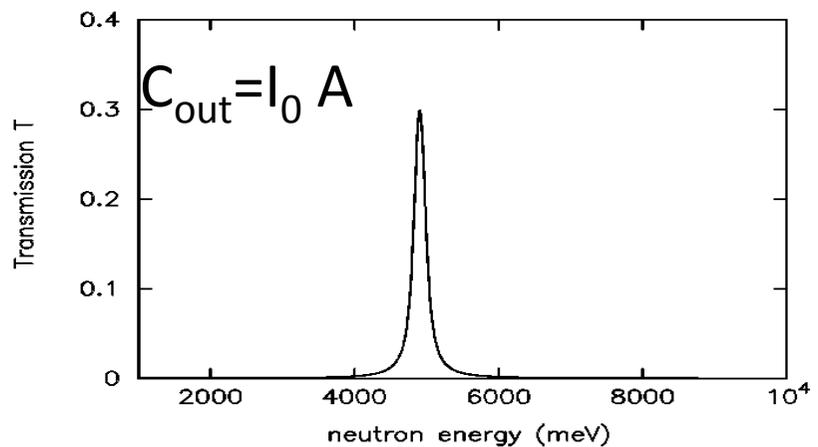
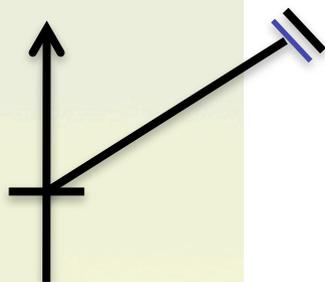


Secondary foils are cycled (50% duty cycle) to remove drifts in detector efficiency with time, due for example to ambient temperature changes.

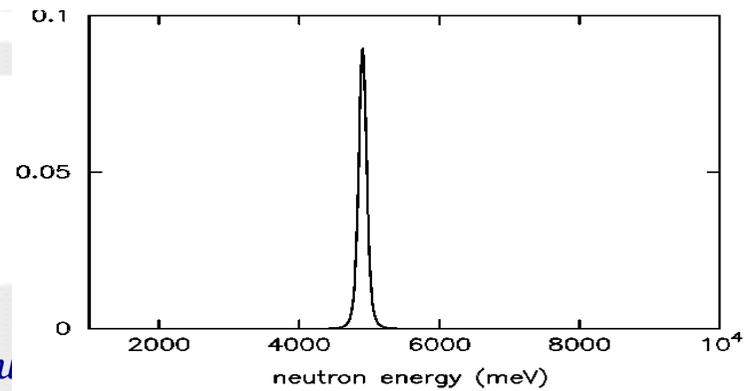
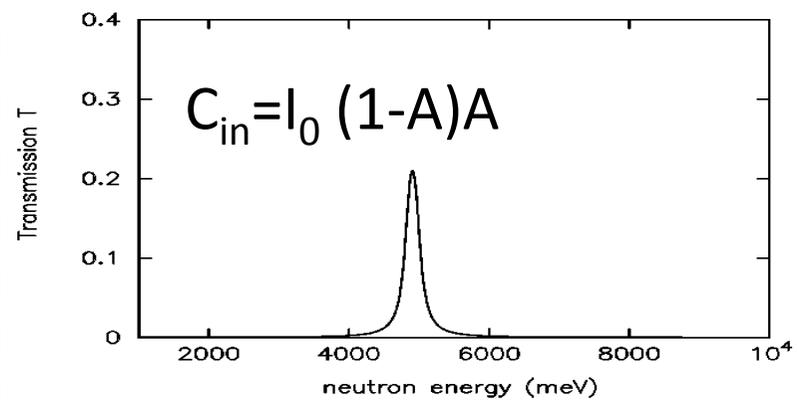
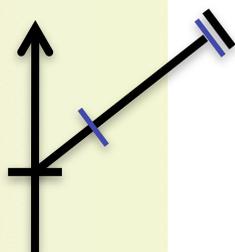
Experimental Tests
at ISIS in 2005



Foil out

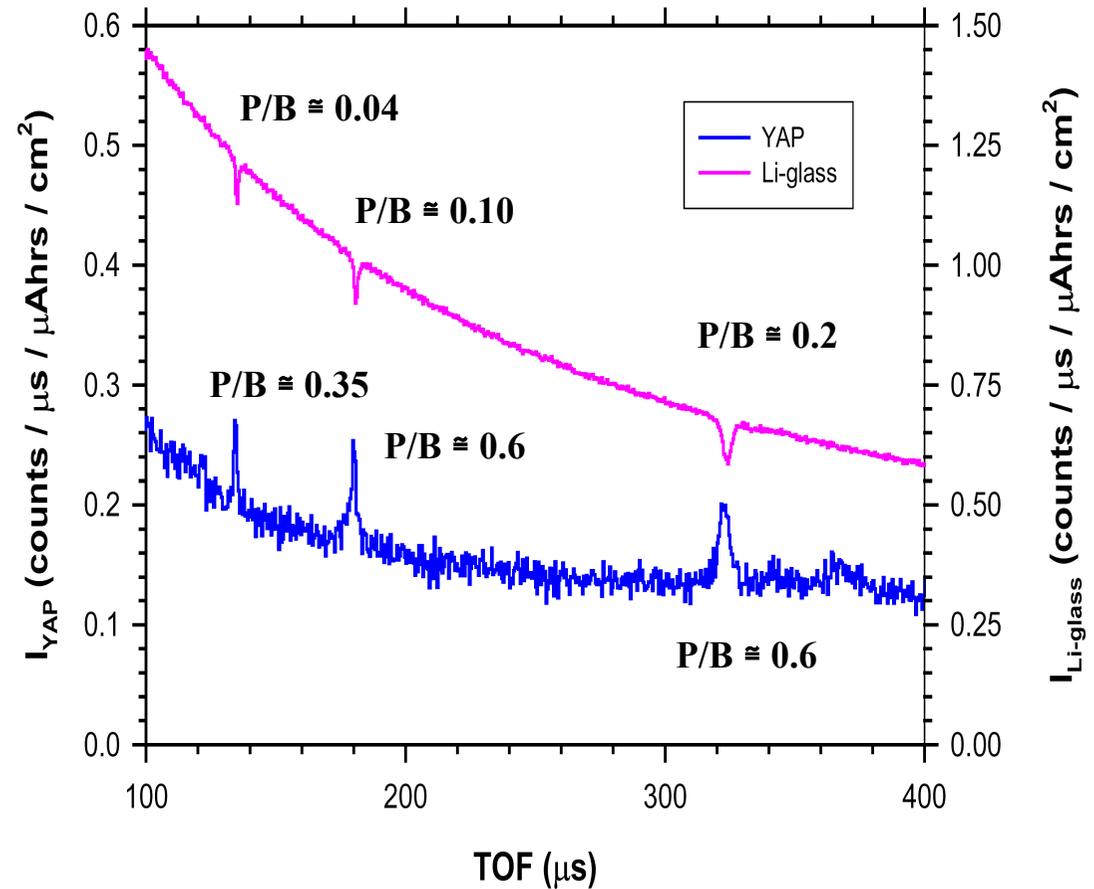


Foil in

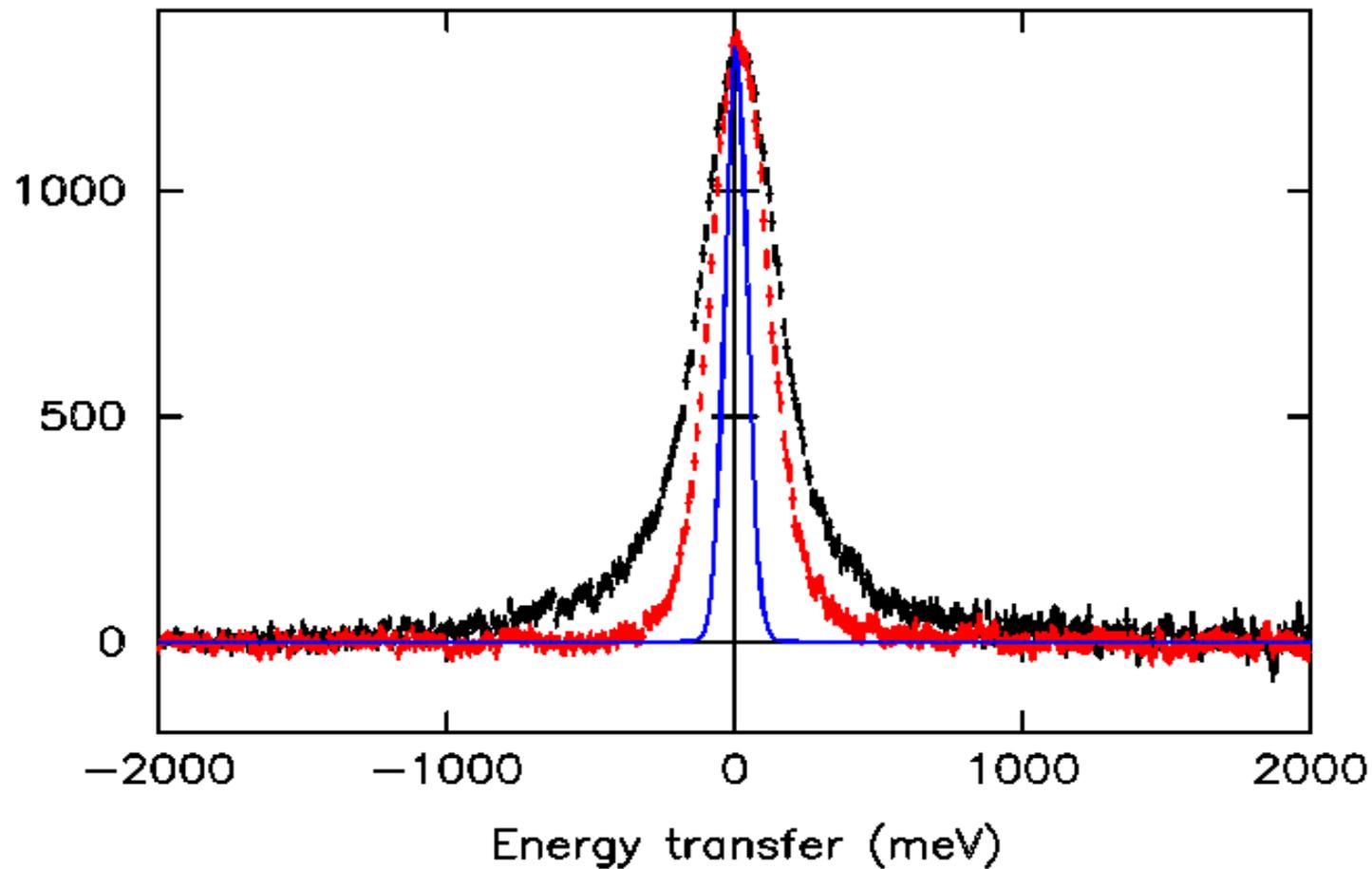


YAP: a comparison with Li-glass

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



Resolution improvement FC versus FD



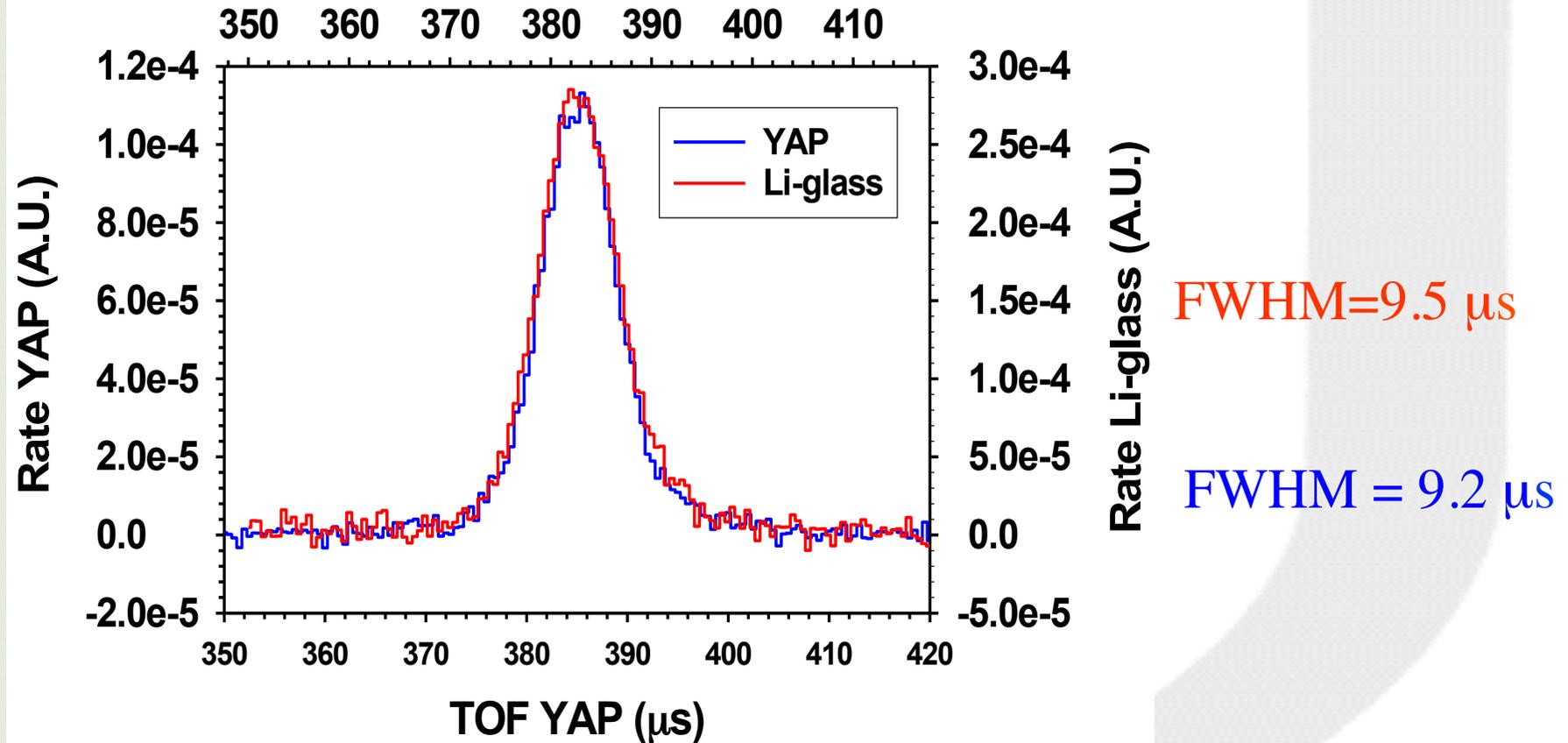
Blue = intrinsic width of lead peak

Black = measurement using Filter Single Difference technique

Red = Foil Cycling technique

Resolution improvement FC versus FDD

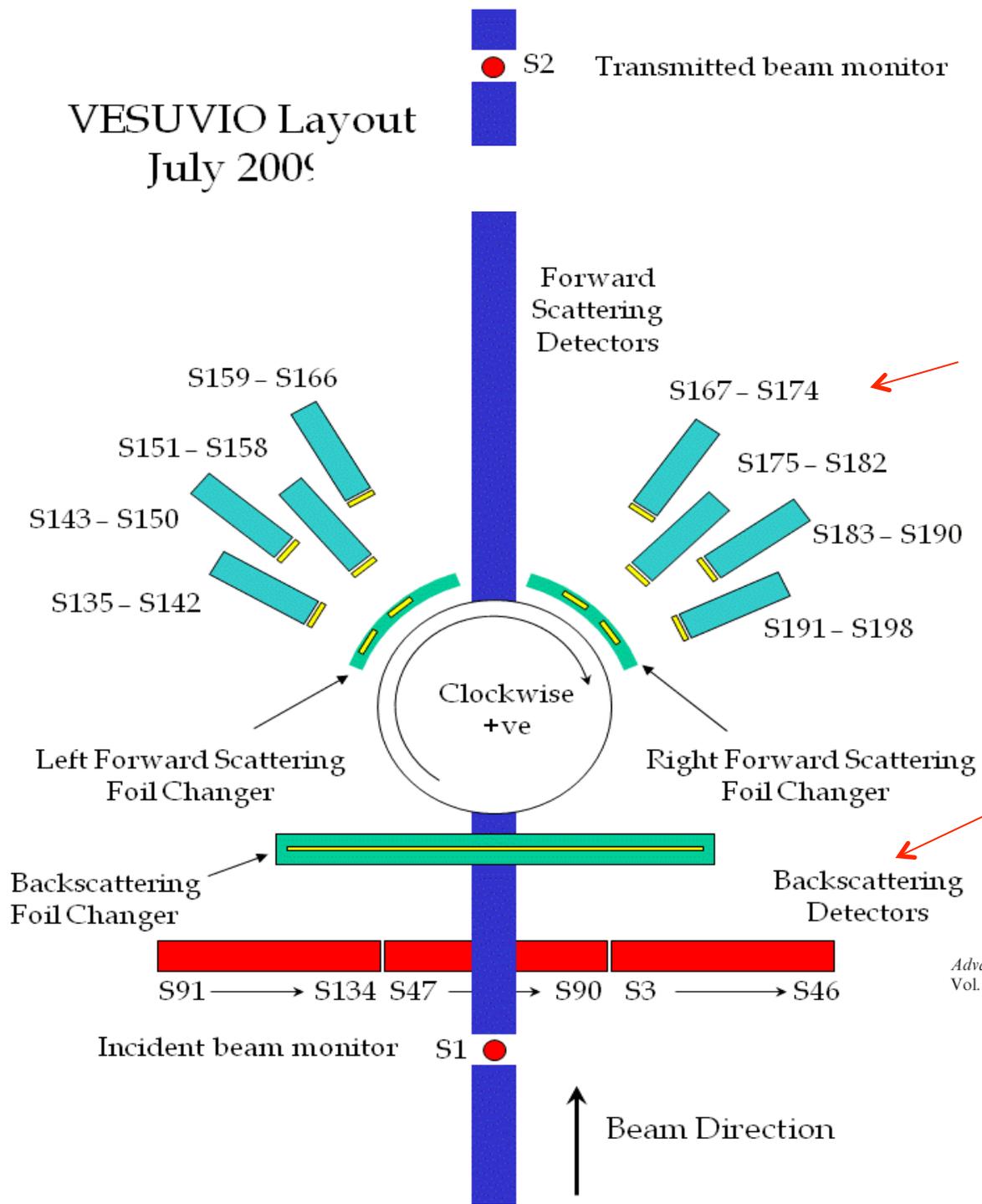
1 mm lead, Li-glass double difference



YAP same resolution as Li-glass in DD

VESUVIO Layout July 2009

VESUVIO from 2006
operates in RDS & RFS



YAP gamma detectors

⁶Li neutron detectors

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



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 (eVS upgrade)

The Resonance Filter Spectrometer (1982-2003)

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

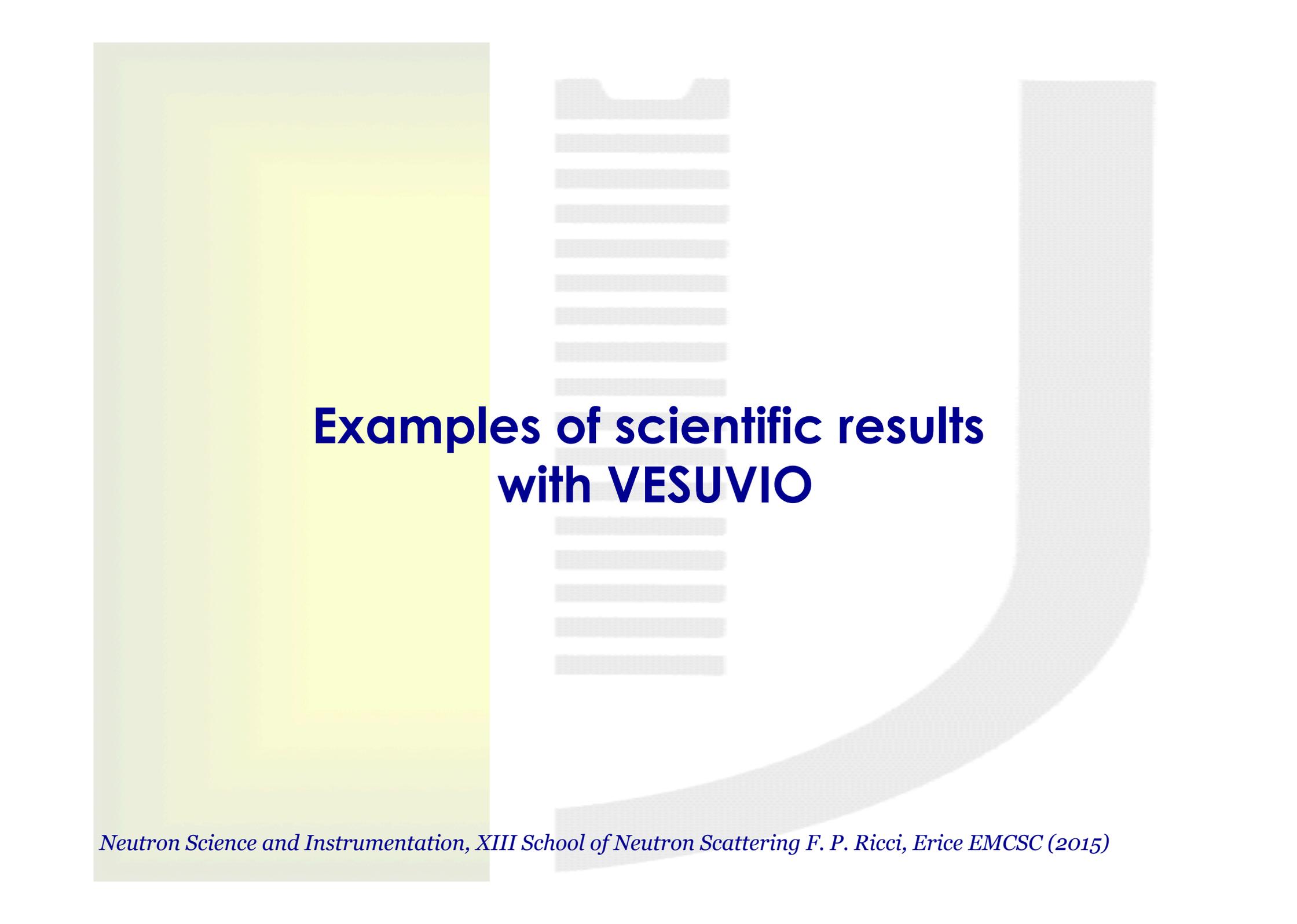
Resonance Detector Spectrometer (from 2002)

- Foil cycling technique (FC)

From 2006:

RDS (YAP detector) & RFS (^6Li neutron detectors)

- Due to installation of YAP detectors VESUVIO has gained one order of magnitude better accuracy for proton measurements
- Accuracy in widths of $n_{\text{H}}(p)$ is $\sim 0.5\%$

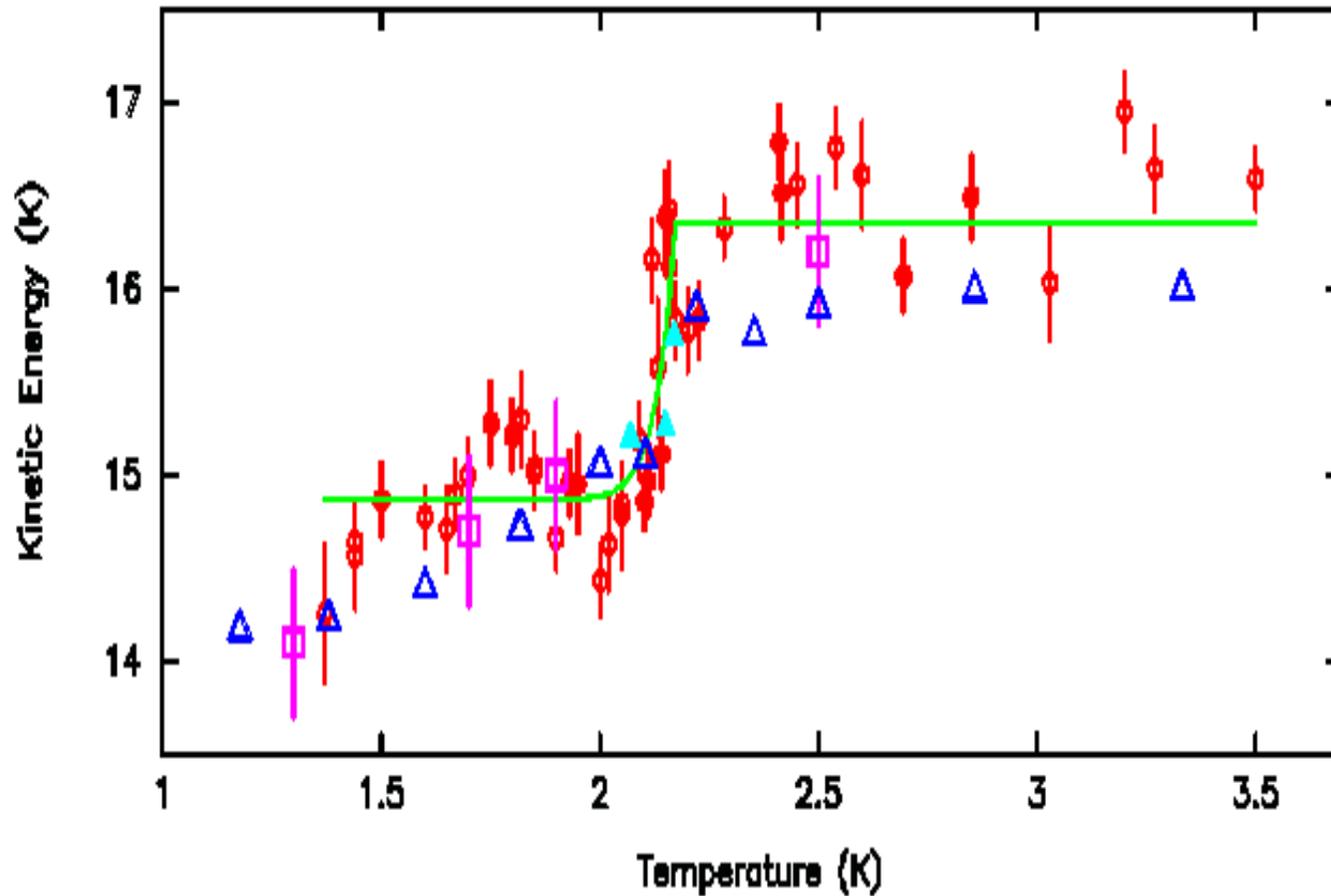
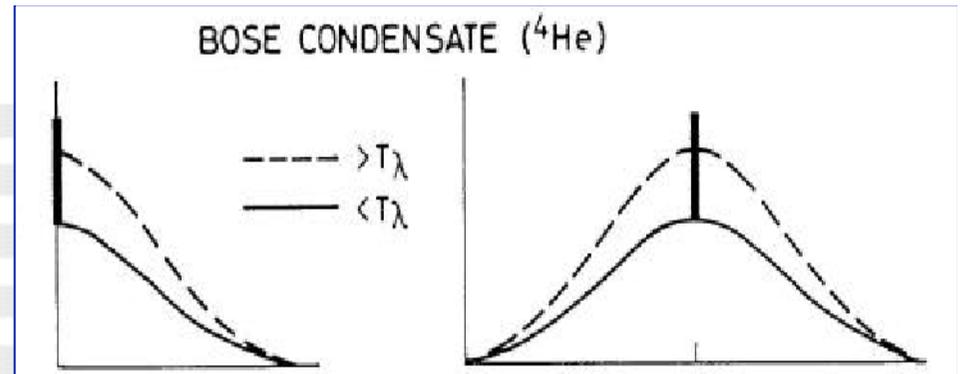


Examples of scientific results with VESUVIO

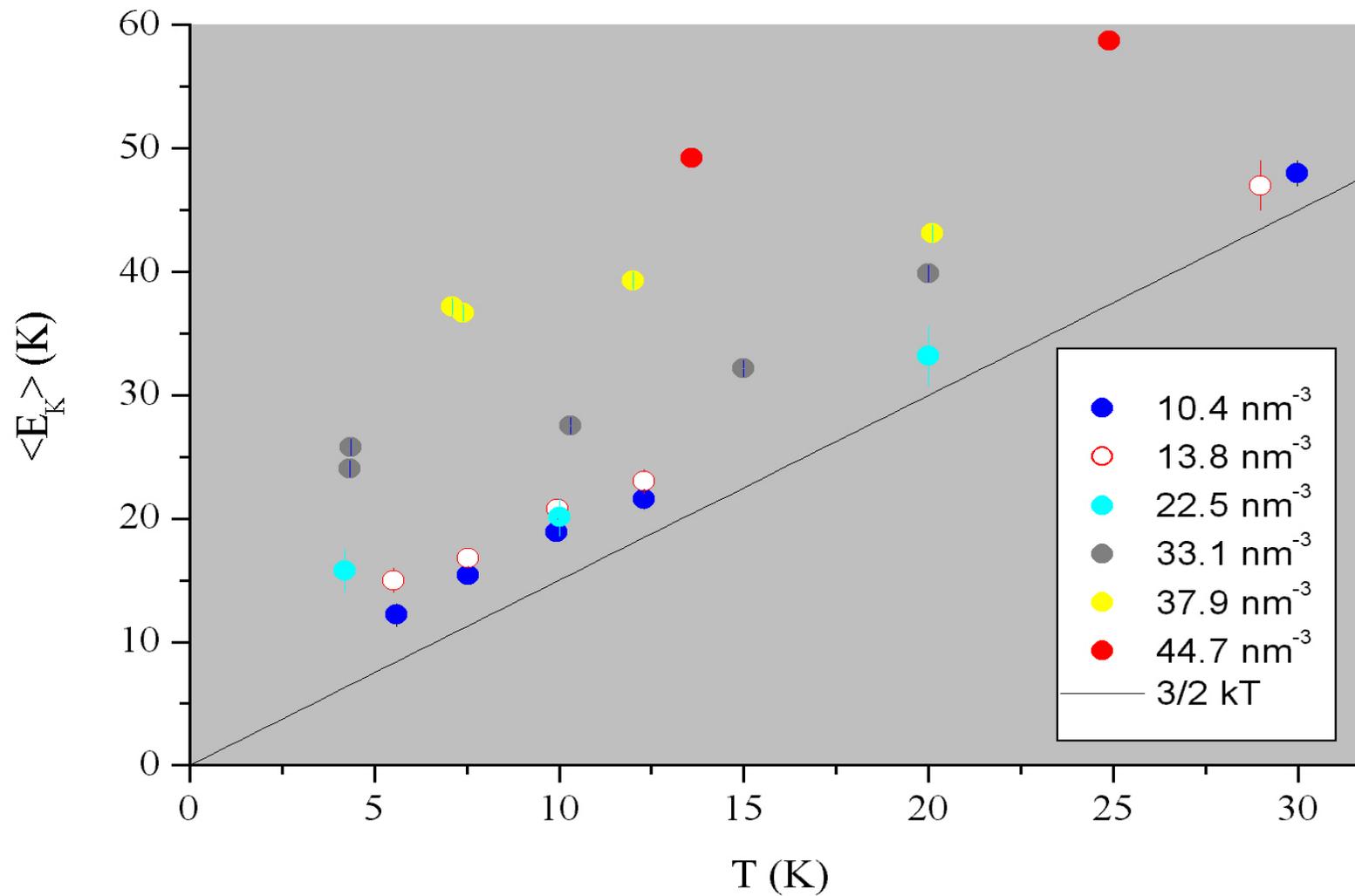
Neutron Science and Instrumentation, XIII School of Neutron Scattering F. P. Ricci, Erice EMCSC (2015)

Mean Kinetic Energy of ^4He

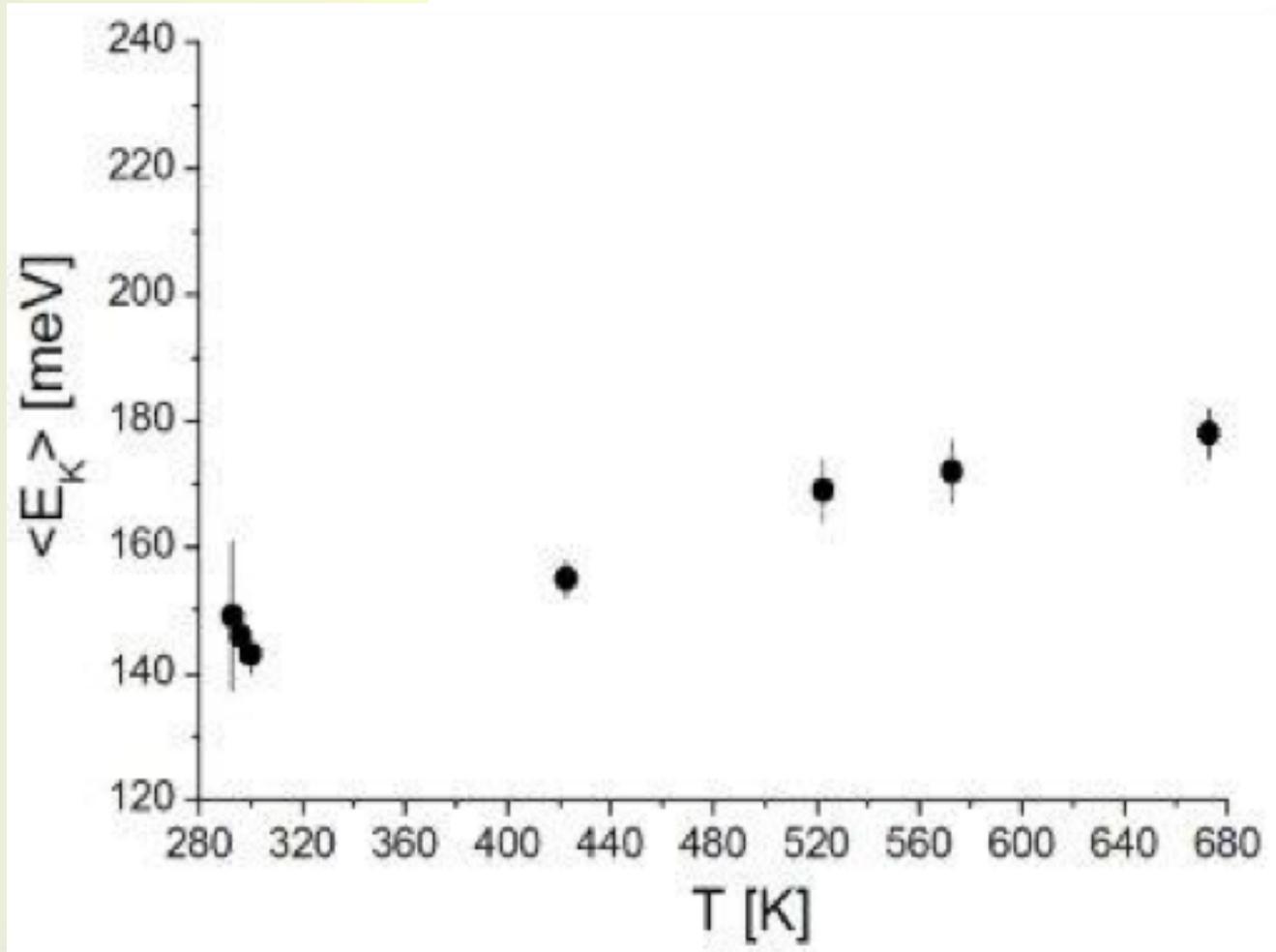
DINS (and INS)



Liquid ^4He $\langle E_K \rangle > 3/2 kT$!



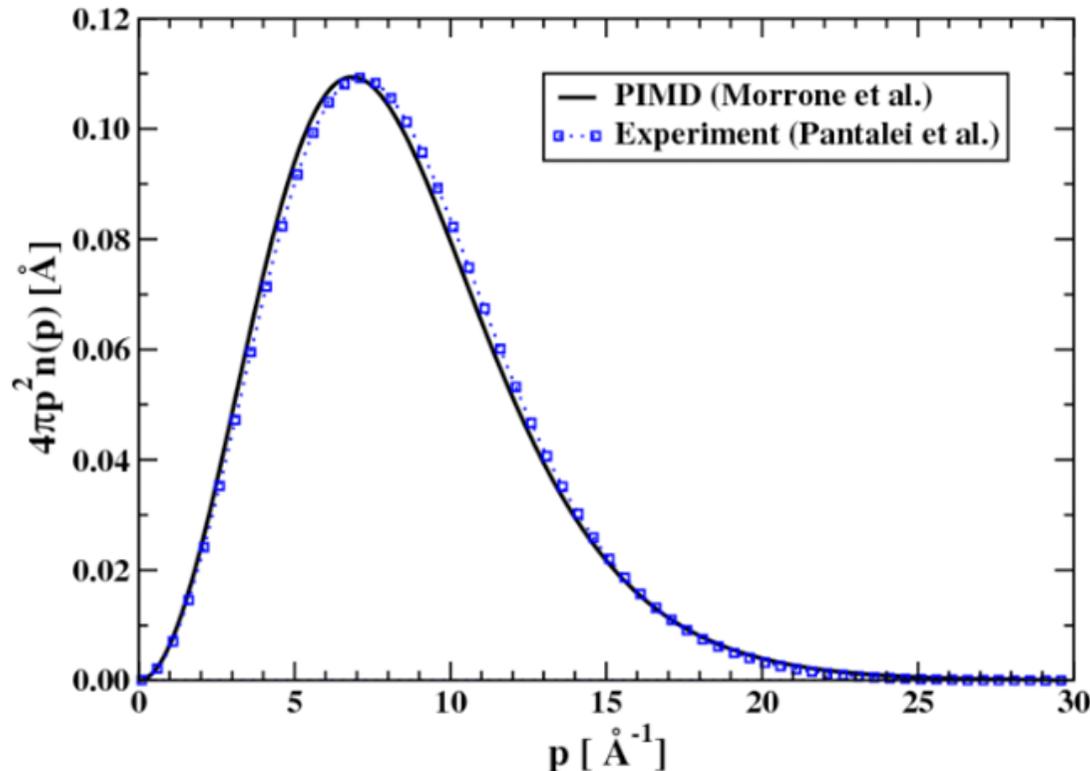
Supercritical water n(p) very similar to the H₂O monomer!



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):

$T = 673 \text{ K}$

$P = 106 \text{ MPa}$

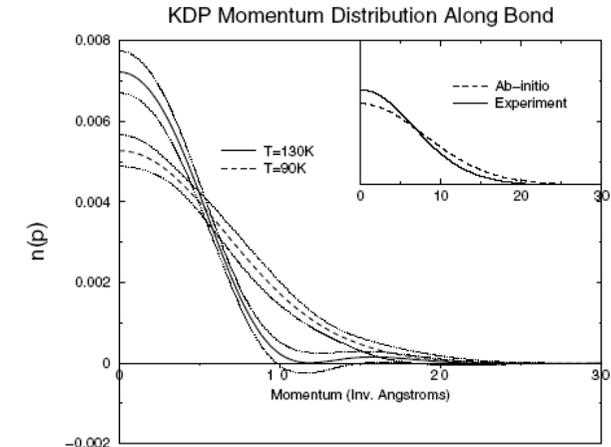
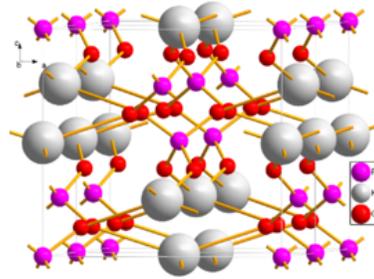
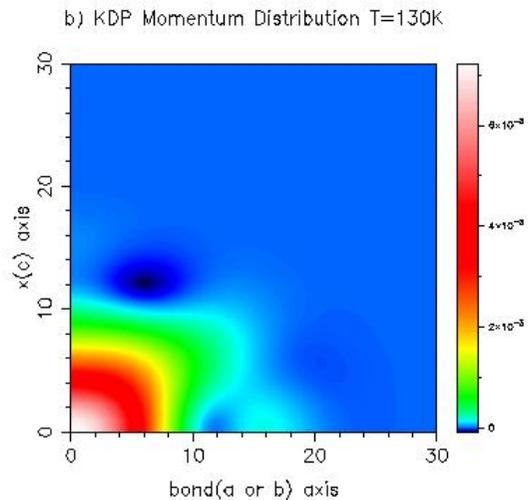
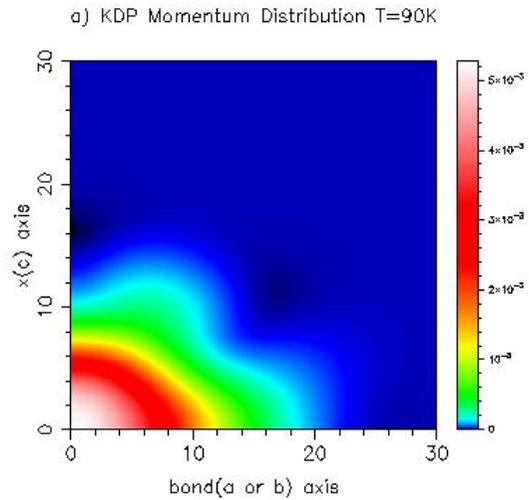
$r = 0.7 \text{ g/cm}^3$

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

$n(p)$ harmonic- anisotropic lineshape :

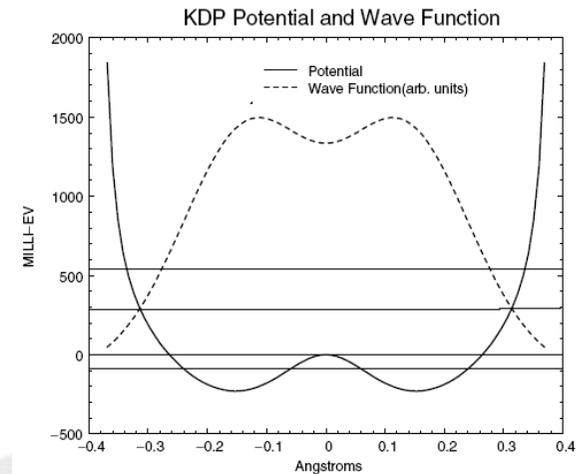
$$n(p) = \left\langle \prod_i \frac{1}{\sqrt{2\pi \sigma_i^2}} \exp\left(-\frac{p^2}{2\sigma_i^2}\right) \right\rangle_{\Omega}$$

High-energy Neutrons as the Ultimate *Wavefunction Diffractometer*



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

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



Requires single crystals

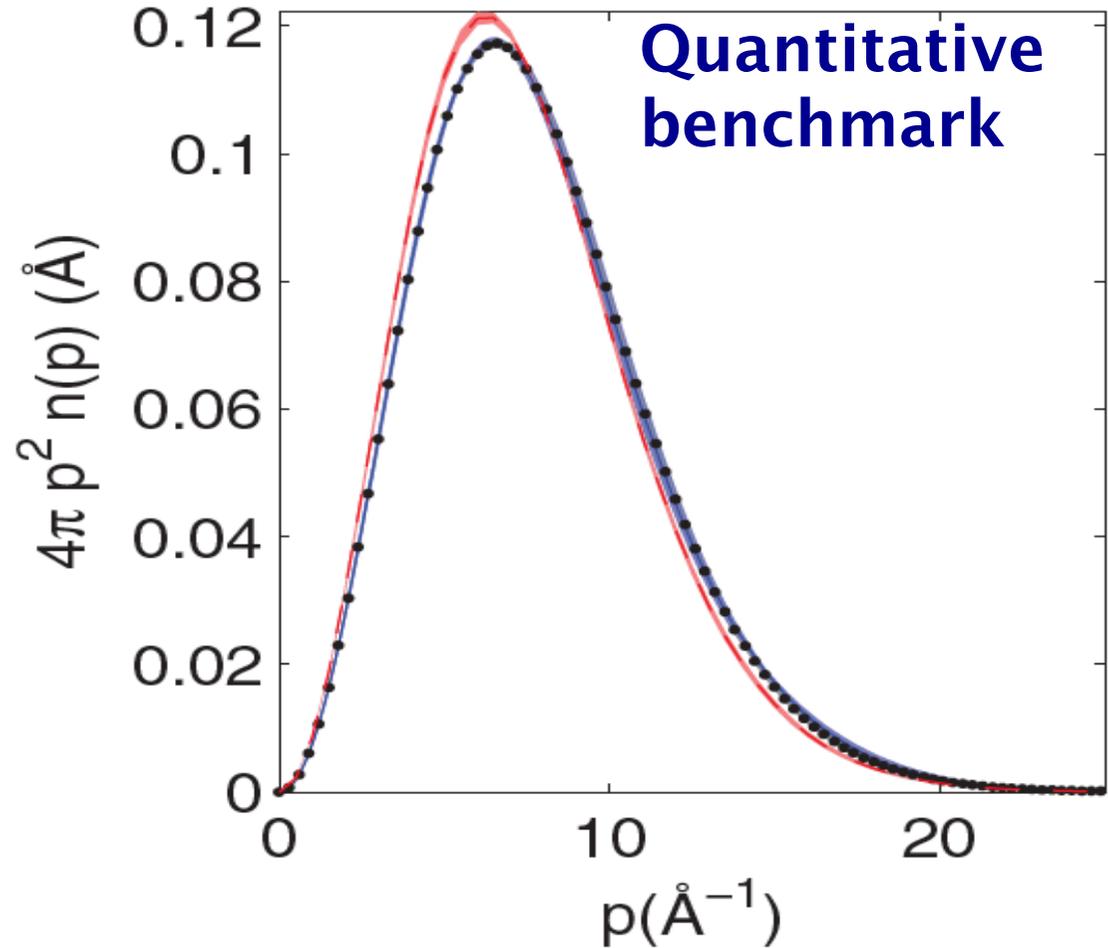


DINS ● at 271 K

$\langle E_K \rangle = 154 \pm 2$ meV

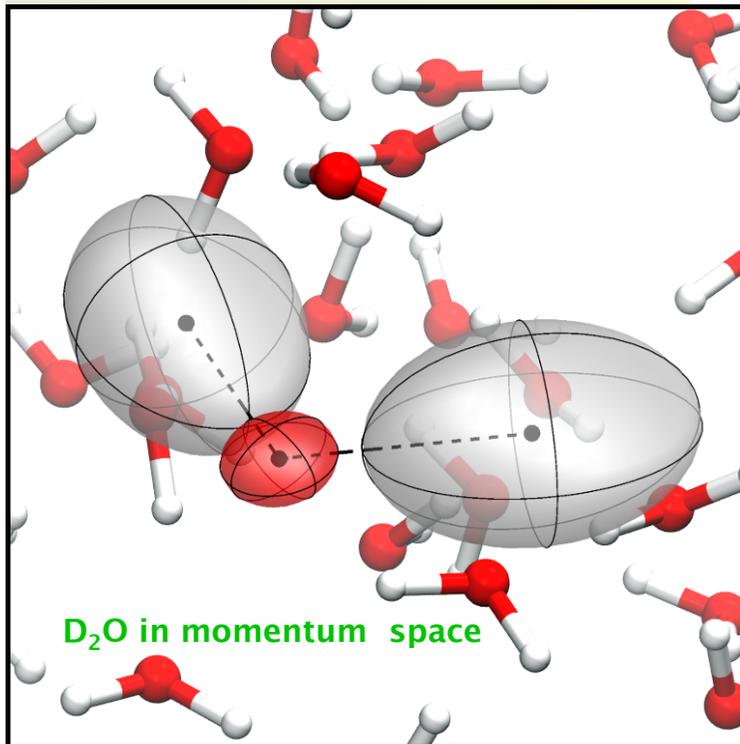
PICPMD ----- at 269 K

$\langle E_K \rangle = 143 \pm 2$ meV

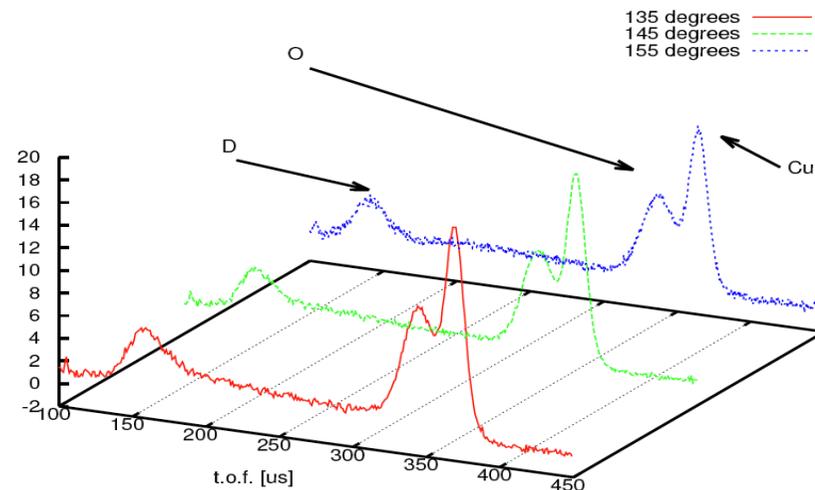


* *D. Flammini, et al., J. Chem. Phys. 136, 024504 (2012)*

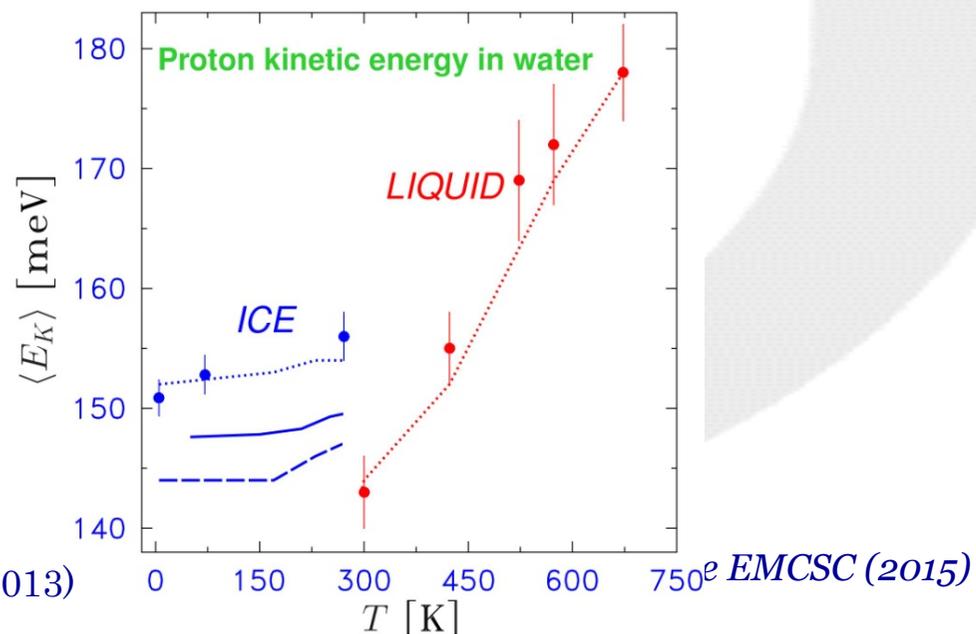
Results from H, D and O in D₂O



R. Senesi, *et al* *Chemical Physics* **427**, 111 (2013)

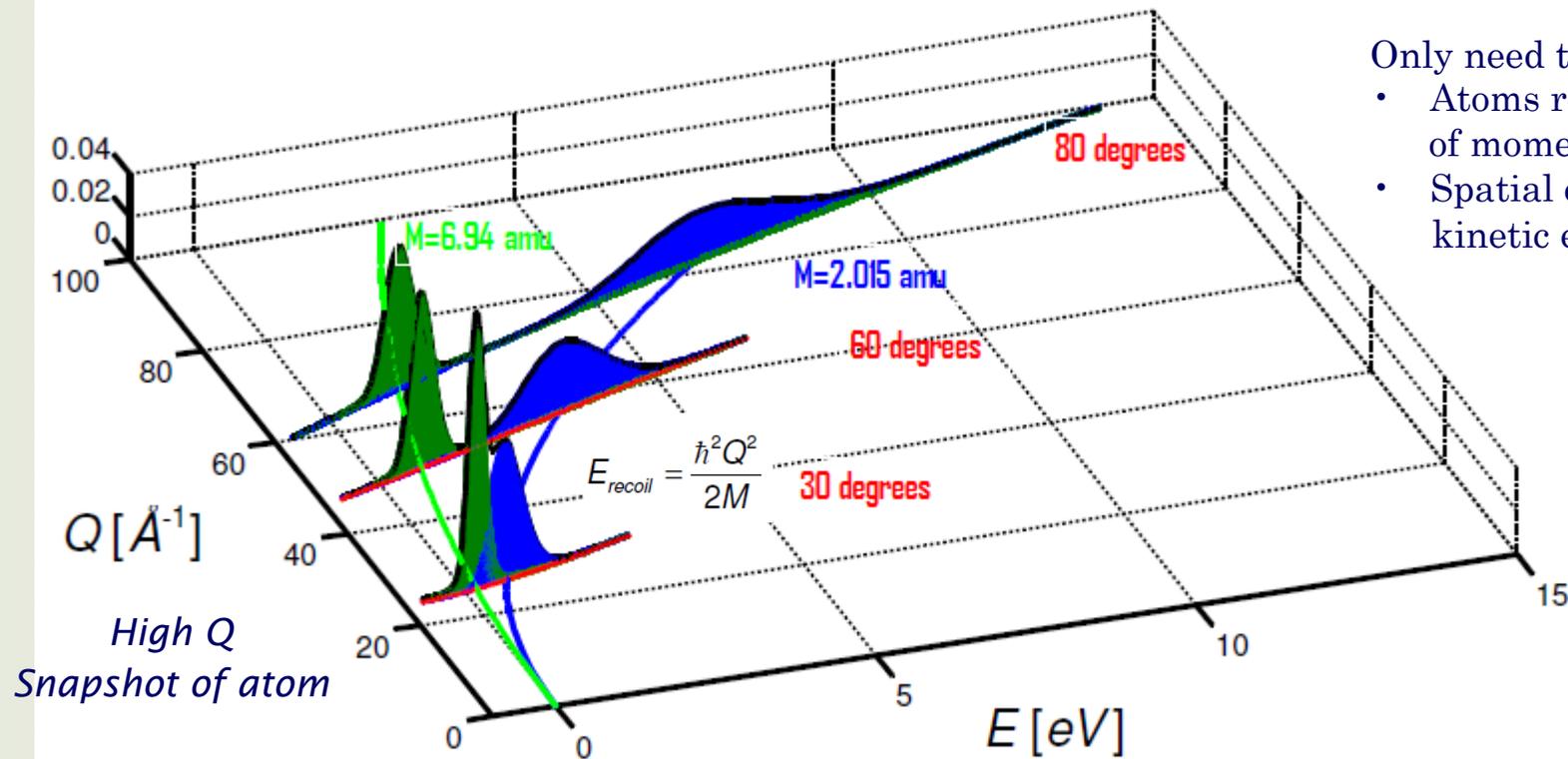


G. Romanelli, *et. al.* *J Phys Chem Lett* **4** 3251 (2013)



EMCSC (2015)

MAss-selective Neutron SpEctroscopy - MANSE



Only need to know that:

- Atoms recoil (conservation of momentum).
- Spatial confinement raises kinetic energy (ZPE).

Atomic Quantum Thermometry:

- Mass selectivity from atomic recoil (kinematics).
- Width of recoil peaks: kinetic energy or 'chemical temperature' of an atom (binding).
- Already demonstrated up to 20 amu.

PHYSICAL REVIEW B 88, 184304 (2013)

Mass-selective neutron spectroscopy of lithium hydride and deuteride: Experimental assessment of the harmonic and impulse approximations

Maciej Krzystyniak* and Selena E. Richards

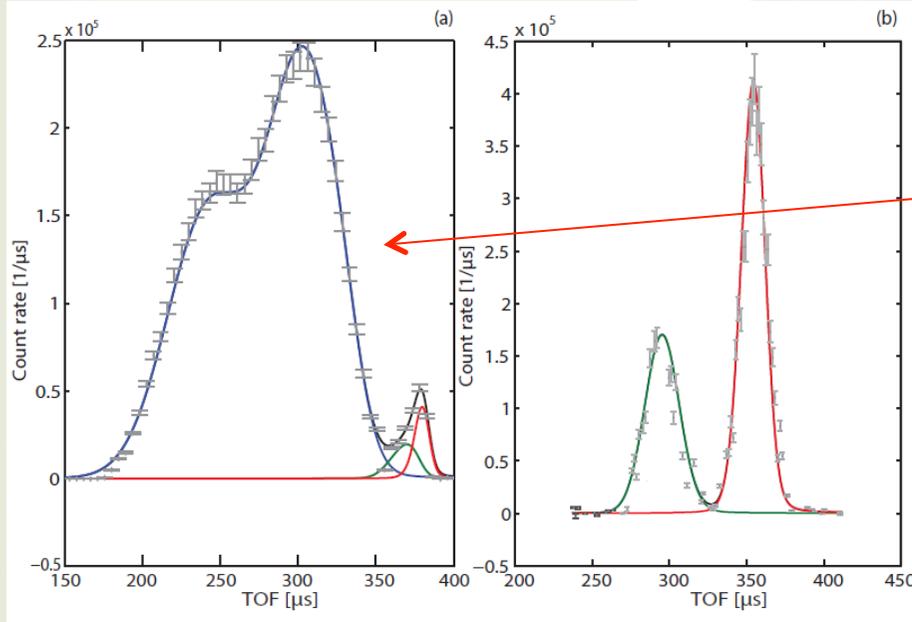
School of Science and Technology, Nottingham Trent University, Clifton campus, Nottingham NG11 8NS, United Kingdom and ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot, Oxfordshire OX11 0QX, United Kingdom

Andrew G. Seel and Felix Fernandez-Alonso†

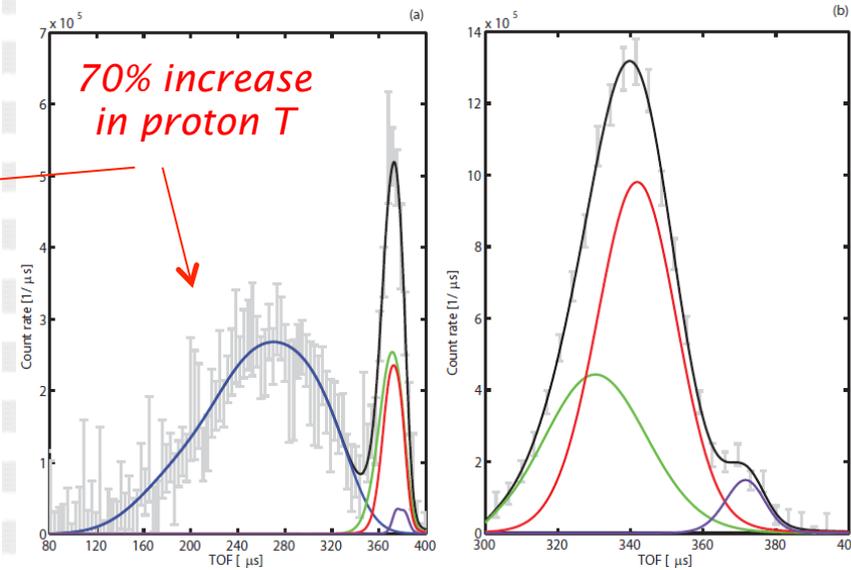
ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot, Oxfordshire OX11 0QX, United Kingdom (Received 26 July 2013; revised manuscript received 22 October 2013; published 19 November 2013)

MANSE: Unique Chemical Information from Recoil Data

❖ Lithium Hydride (ionic)



❖ Squaric Acid (H-bonded)



- ❖ Two modes of operation (similar to xtallography):
 - ❖ Coarse resolution (forward scattering, H)
 - ❖ High resolution (backscattering)
- ❖ Peak integration: head count with sub-ppm sensitivity for H.
- ❖ Sensitive to chemical environment (temperature) around an atom, a consequence of binding forces and dimensionality of bonding network.
- ❖ Mass resolution could be improved further.

(in press)

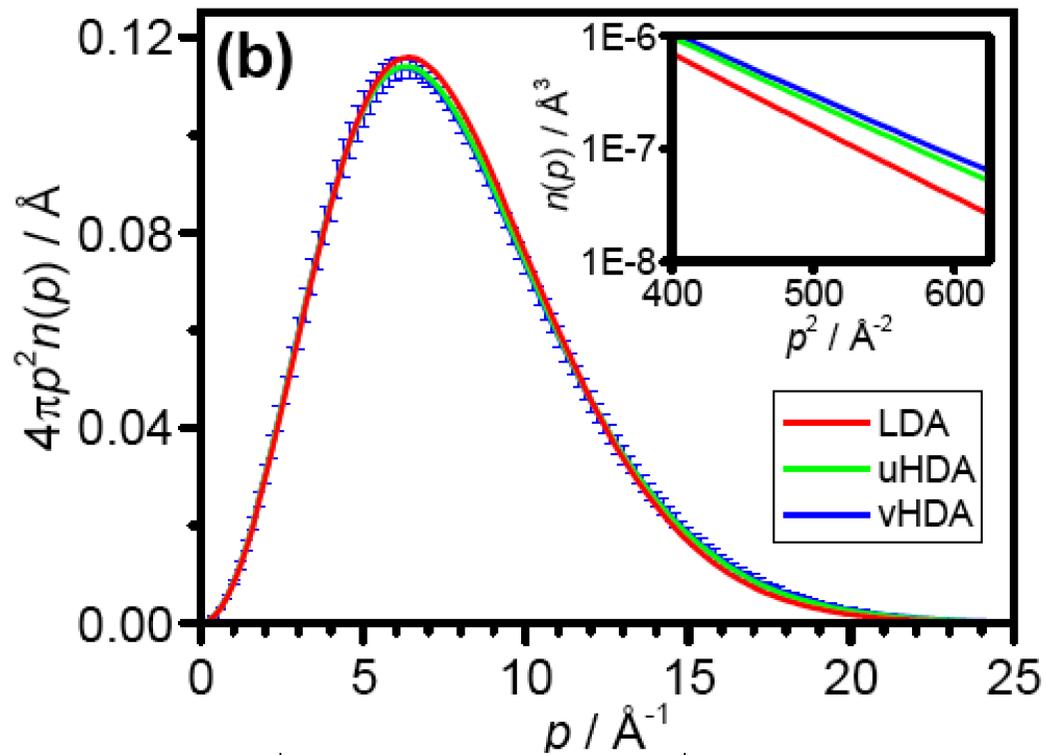
Mass-selective Neutron Spectroscopy Beyond the Proton

M. Krzystyniak^{1,2}, A.G. Seel¹, S.E. Richards^{1,2}, M.J Gutmann¹, and F. Fernandez-Alonso^{1,3}

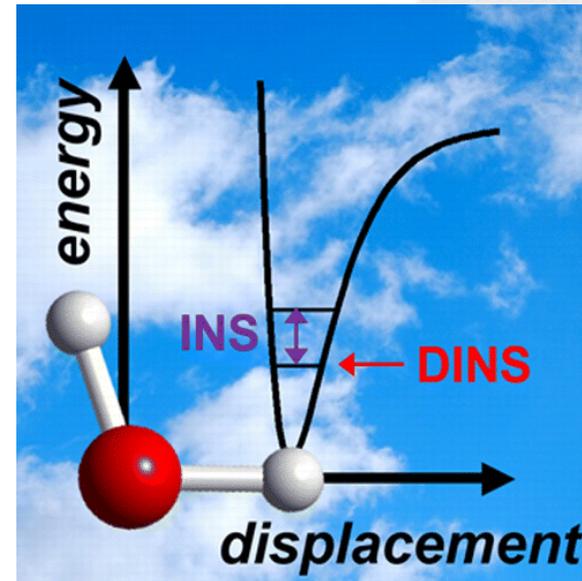
¹ ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot, Oxfordshire OX11 0QX, United Kingdom

² School of Science and Technology, Nottingham Trent University, Clifton Campus, Nottingham NG11 8NS, United Kingdom

³ Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, United Kingdom



AMORPHOUS ICE

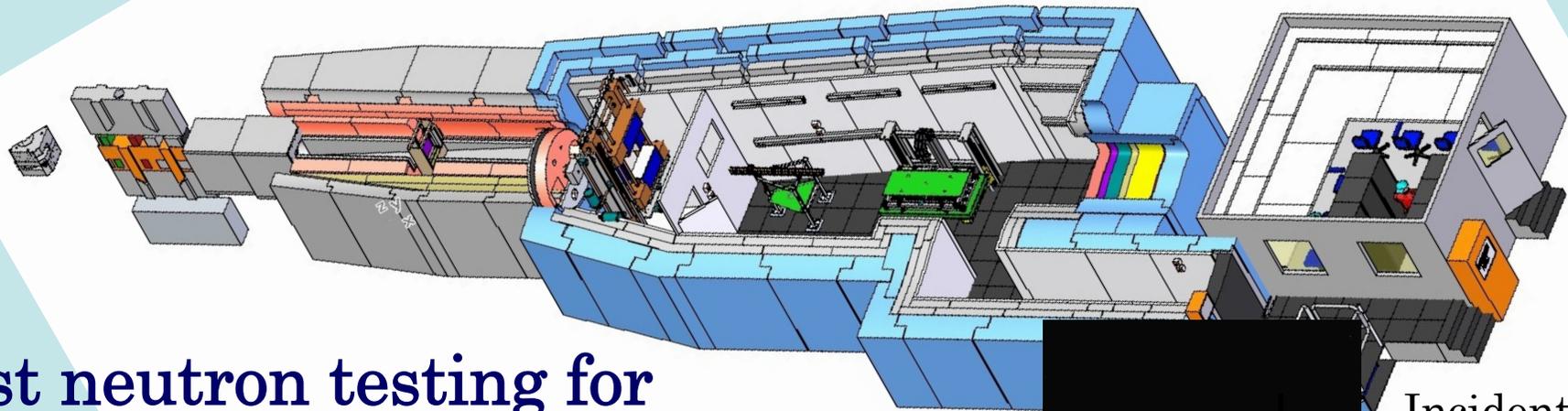


	Ice Ih		LDA		uHDA		vHDA	
$\rho / \text{g cm}^{-3}$	0.934		0.94		1.15		1.25	
T / K	71	80	80		80		80	
	DINS	INS	DINS	INS	DINS	INS	DINS	INS
$\langle E_K \rangle / \text{meV}$	152.8±1.6	153±3	152±3	151±2	157±3	152±2	159±3	151±2
$\langle E_K \rangle_x / \text{meV}$	-	20.5±1	19.5±2	18.8±1	18.5±2	17.9±1	18.7±1	17.8±1
$\langle E_K \rangle_y / \text{meV}$	-	33.4±1	49.6±3	33.4±1	44.1±3	33.1±1	41.4±1	33.0±1
$\langle E_K \rangle_z / \text{meV}$	-	98.9±1	83.3±6	99.2±1	94.8±5	100±1	98.7±3	101±1

A. Parmentier et al. J. Phys. Chem. Lett. 6, 2038–2042 (2015)

Fig. 1.1. Ricci, Direct DYNOS (2015)

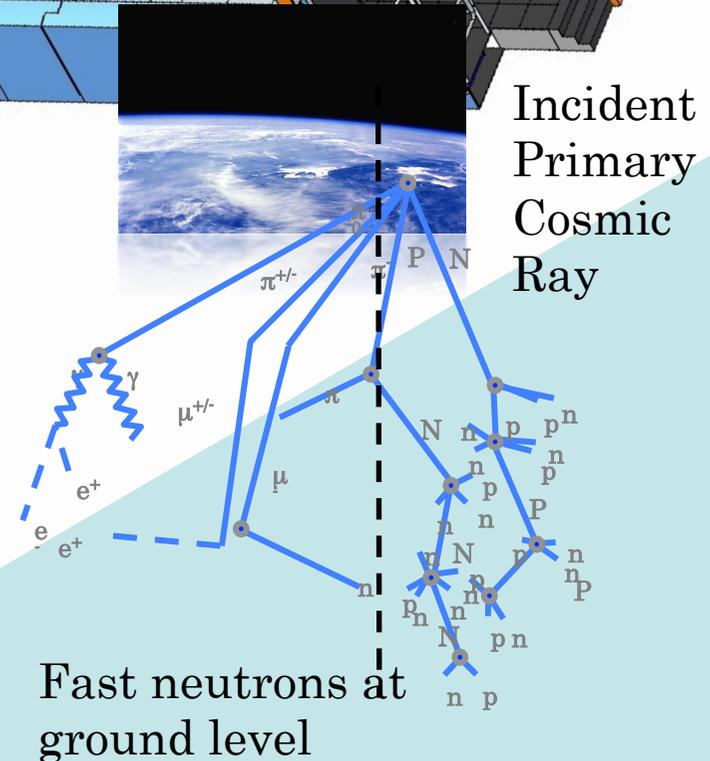
R&D with eV (MeV) neutrons...has triggered new technology and applications at ISIS:
Cultural Heritage (2006: ANCIENT CHARM project)
Chip Irradiation: (2006: CHIPIR)



Fast neutron testing for the semiconductor industry

Chip-ir

At ISIS we are able to simulate Single Events Effects at an accelerated rate so that one hour in the Chipir beamline will be equivalent to a hundred years of aircraft flying time.



Something we did not talk about

Data corrections for DINS measurements

- ❖ Gamma background: Measurements/corrections
- ❖ Multiple scattering: Measurements/corrections

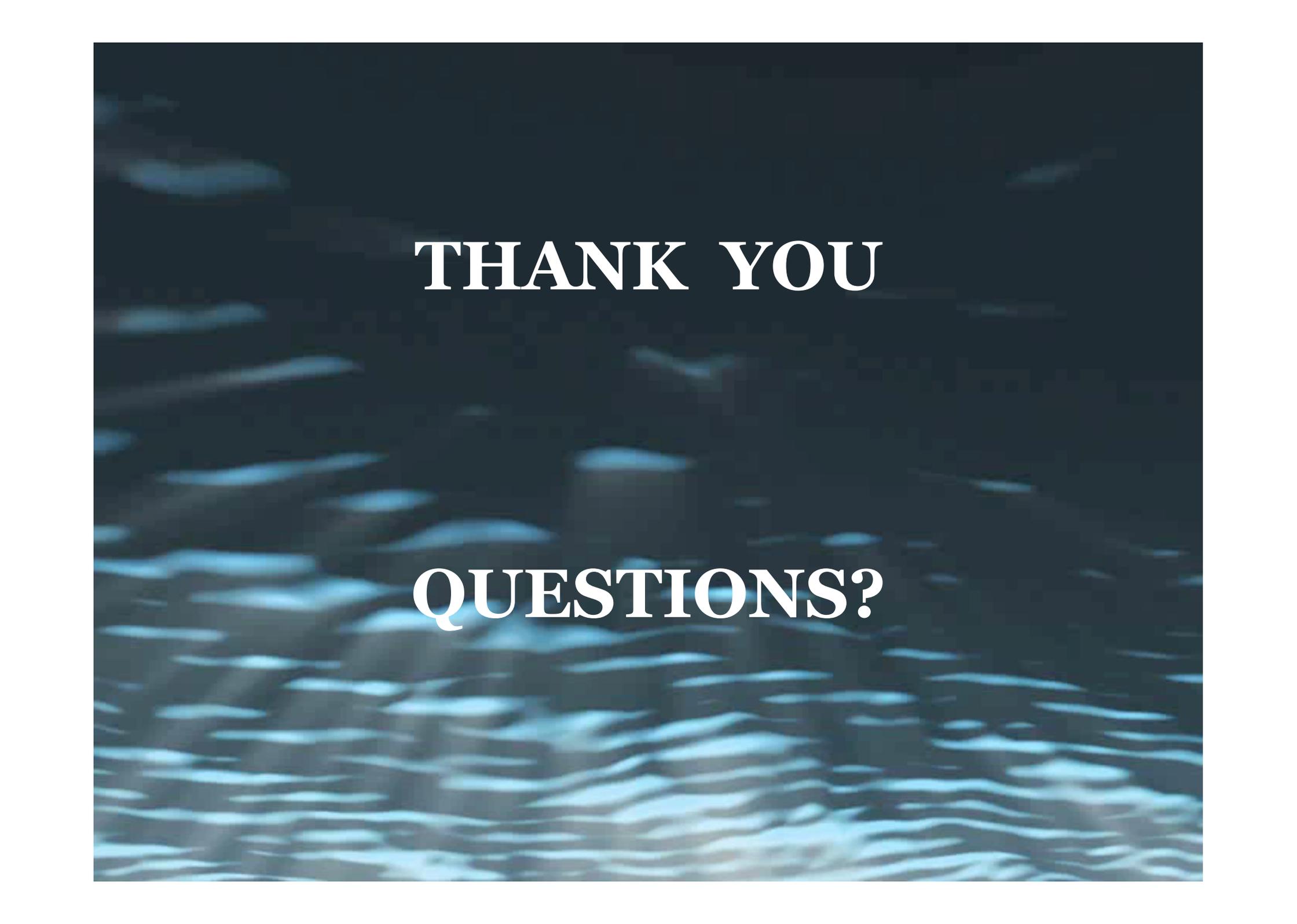
Filter Spectrometers for eV neutron spectroscopy at low q and high w

- ❖ What do we measure
- ❖ Resolution components

Future development

Resonance filter spectrometers in Direct Geometry - complementary use of VESUVIO-like and Chopper spectrometers for $n(p)$

DINS from polyatomic systems, $N(P)$ and $n(p)$

The background of the slide is a close-up, slightly blurred image of blue water with gentle ripples. The lighting is soft, creating a calm and serene atmosphere. The text is centered and rendered in a white, serif font.

THANK YOU

QUESTIONS?