A Guide to HFBS Experiment

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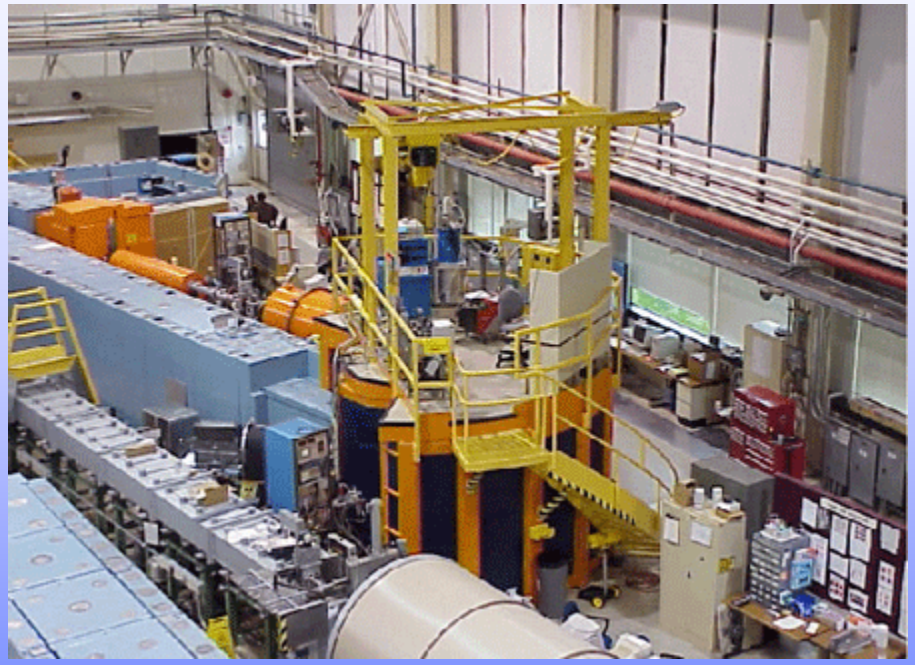
High Flux Backscattering Instrument



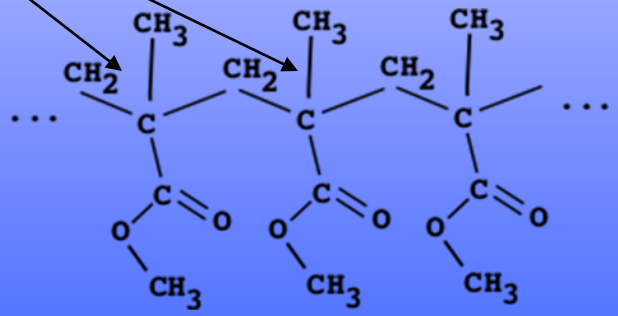
Why Neutrons?

Wavelength of neutrons:

$$\lambda \approx \frac{9}{\sqrt{E}}$$

At 2meV, wavelength of neutrons is about 5Å → typical length scale of interest to researchers and industries

length scales explored: in terms of Q



Poly(methyl methacrylate)

Measurable Quantity in Neutron Scattering

Intensity of scattered neutrons in a given direction θ:

$$\frac{d\sigma(\theta)}{d\Omega dE} = \frac{k_f}{k_i}\frac{1}{N}\sum_{i,j} < b_i b_j > S_{i,j}(Q,\omega)$$

Intermediate scattering function S(Q,ω) or S(Q,t) depends only on your sample!

DSF and ISF are related by Fourier transform.

$$S(Q,\omega) = \int_{-\infty}^{\infty} S(Q,t)e^{-i\omega t}dt$$

$$S(Q,t) = \int_{-\infty}^{\infty} S(Q,\omega)e^{i\omega t}d\omega$$

Intermediate scattering functions

Incoherent

atom-atom self correlation

$$S^{self}{}_i(Q,t) = \left\langle \sum e^{iQ.\left[R_n^i(t) - R_n^j(0)\right]} \right\rangle$$

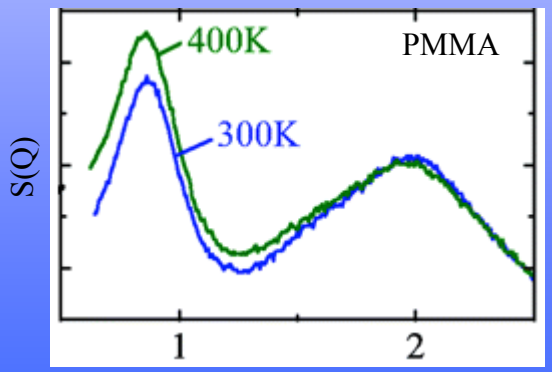
Incoherent intermediate scattering function relates the motion of a given nucleus at t=0 and at a later time t;

Coherent

pair correlation function

$$S_{ij}(Q,t) = \left\langle \sum_{n,m} e^{iQ.\left[R_n^i(t) - R_m^j(0)\right]} \right\rangle$$

Coherent scattering function relates position of a pair of atoms at different times



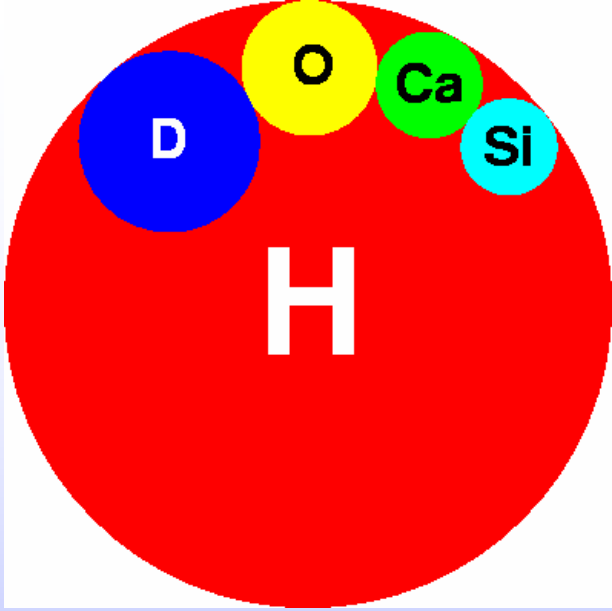
time integral

$$S(Q,\omega) = FT * S(Q,t)$$

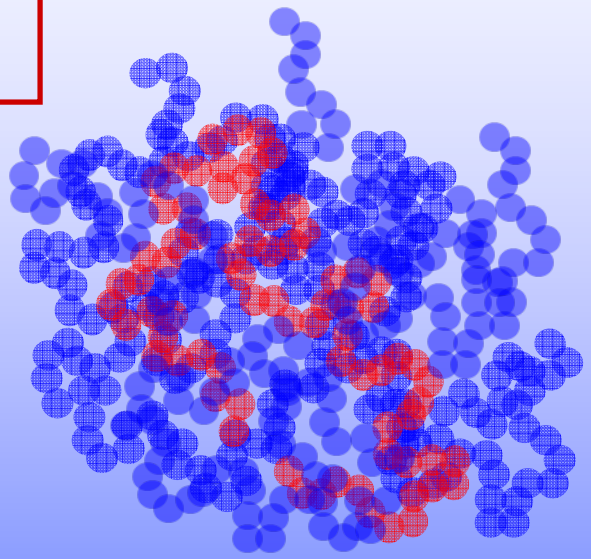
Importance of hydrogen?

Scattering cross sections

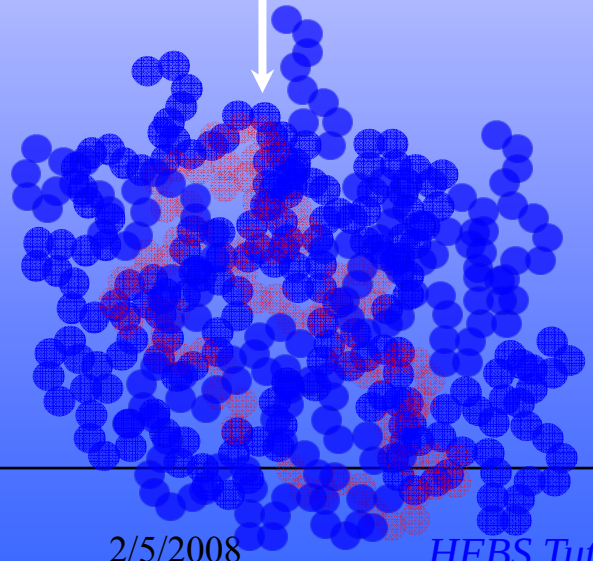
elements	Incoherent	coherent
σ (H) ~	82	2
σ (D) ~	2	5
σ (C) ~	0	5
σ (O) ~	0	4

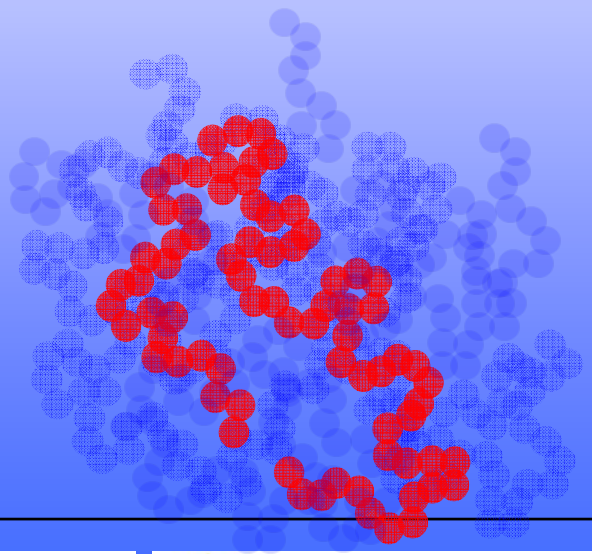


A(H)/B(D)



A(D)/B(H)





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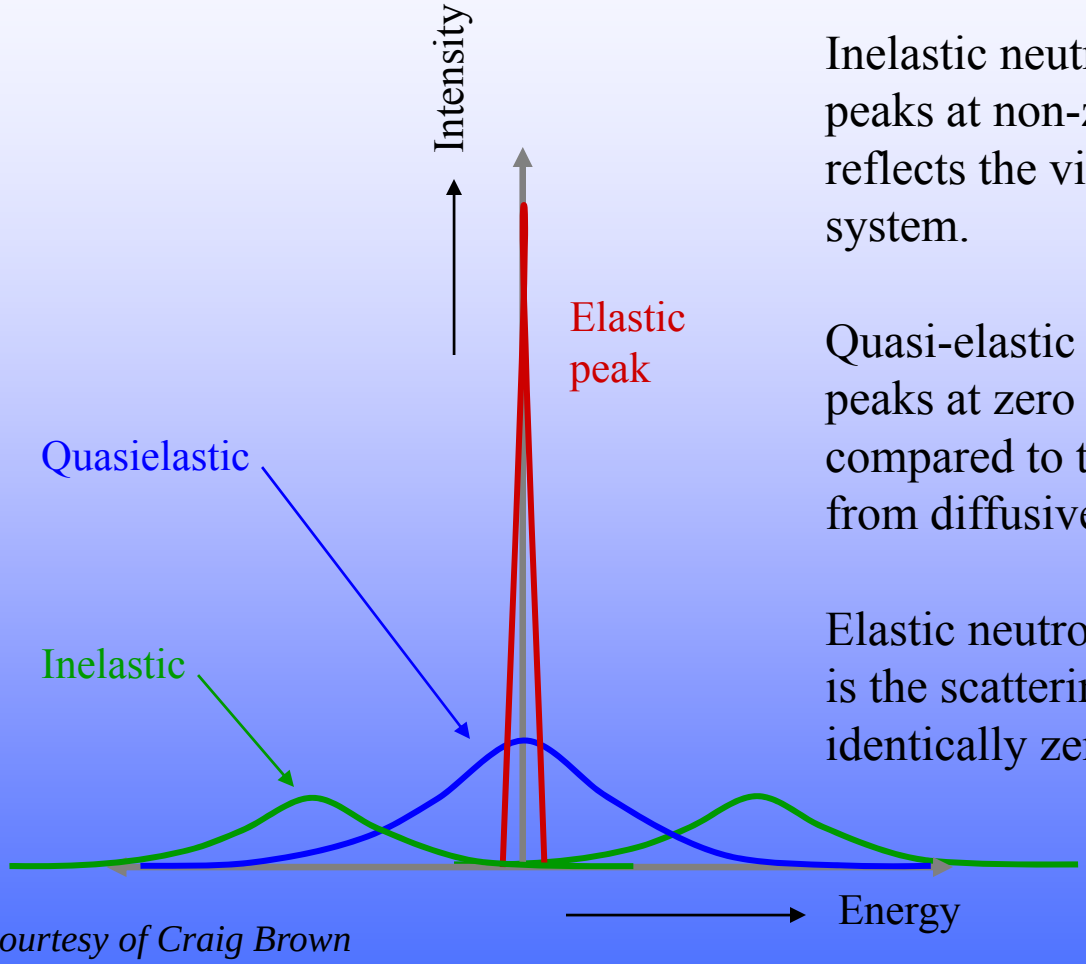
Quasi-elastic and inelastic Neutron scattering



Courtesy of Craig Brown

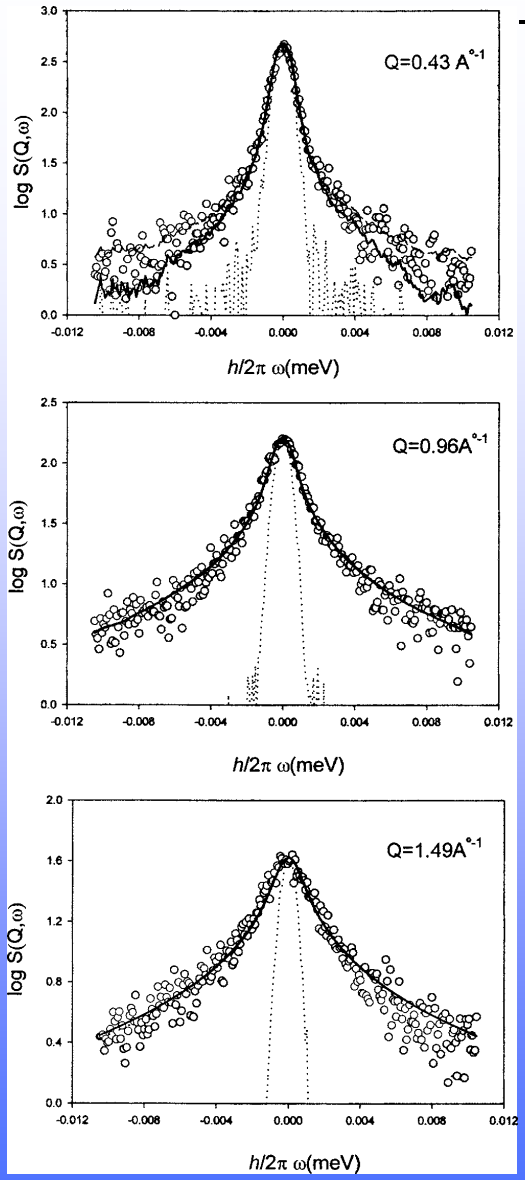
Inelastic neutron scattering:
peaks at non-zero energy transfer. This scattering reflects the vibrational or fast modes of the system.

Quasi-elastic neutron scattering:
peaks at zero energy transfer, but is broadened compared to the instrumental resolution. It arises from diffusive or diffusive-like processes.

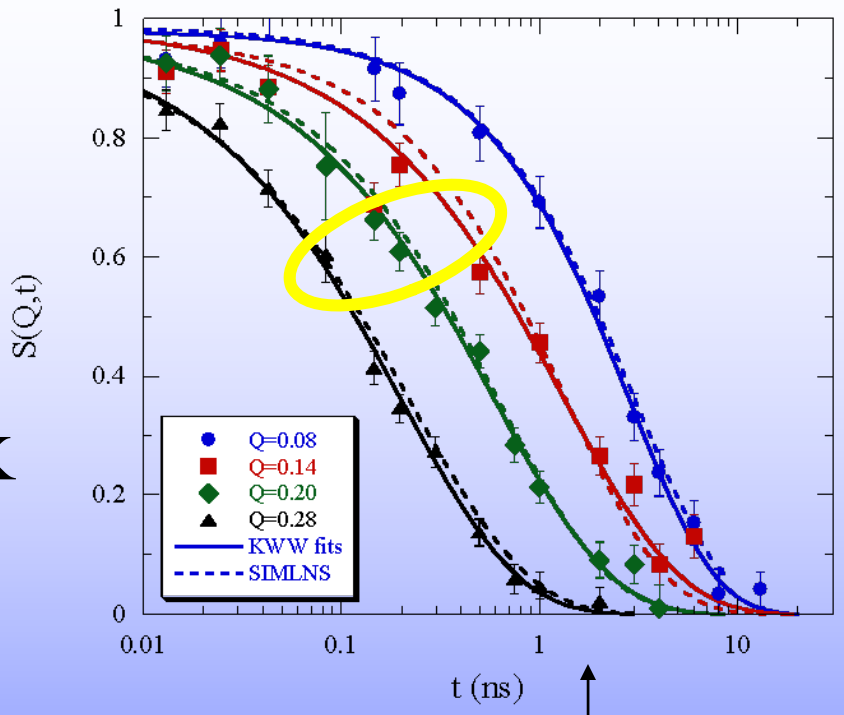
Elastic neutron scattering
is the scattering for which the energy transfer is identically zero.

Time domain Vs frequency domain

BS



NSE



PVAc at 460K

$$S(Q,t) = A(Q)\ \exp\left[-\left(\frac{t}{\tau_w}\right)^{\beta}\right]$$

$$S(Q,\omega) = A(Q) * FT\left\{\exp\left[-\left(\frac{t}{\tau_w}\right)^{\beta}\right]\right\} \Leftarrow \beta=0.50$$

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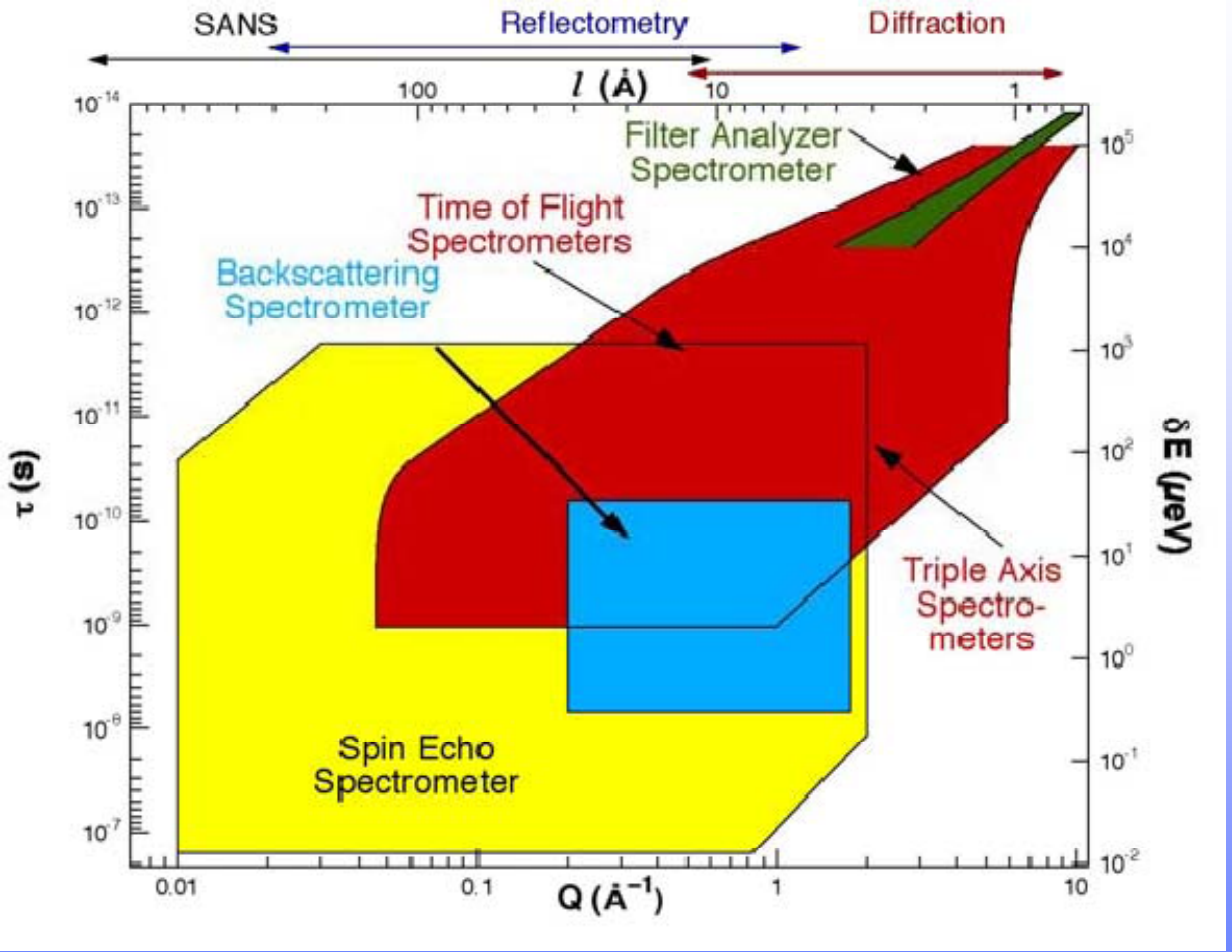
Dynamics and Neutron Backscattering

7 orders of magnitude in energy and 3 orders in length scales!

The range covered by HFBS makes it most suitable for the dynamics of polymers, small molecules and biological systems.



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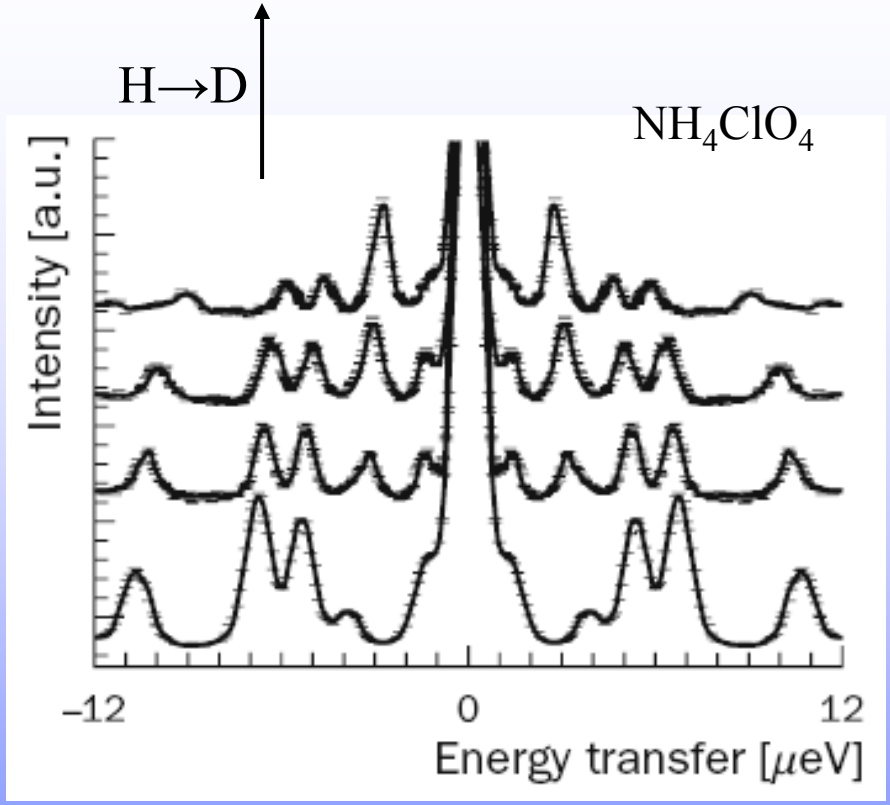
Why Backscattering?

Basic eq. for resolution:

$$\frac{\delta\lambda}{\lambda} = \frac{\delta d}{d} + \frac{\delta\theta}{\tan\theta}$$

intrinsic term depends on crystal properties only



depends only on angular divergence

minimized in BS by choosing θ to be 90 degrees!

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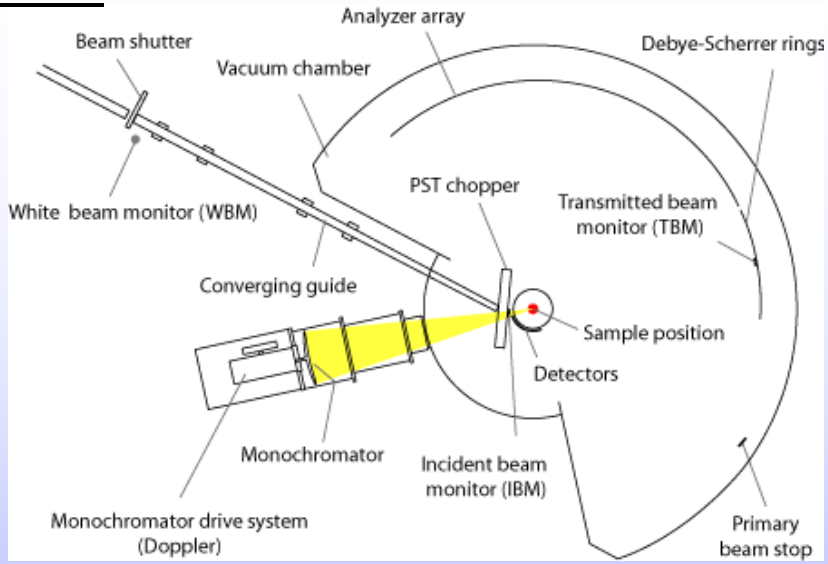
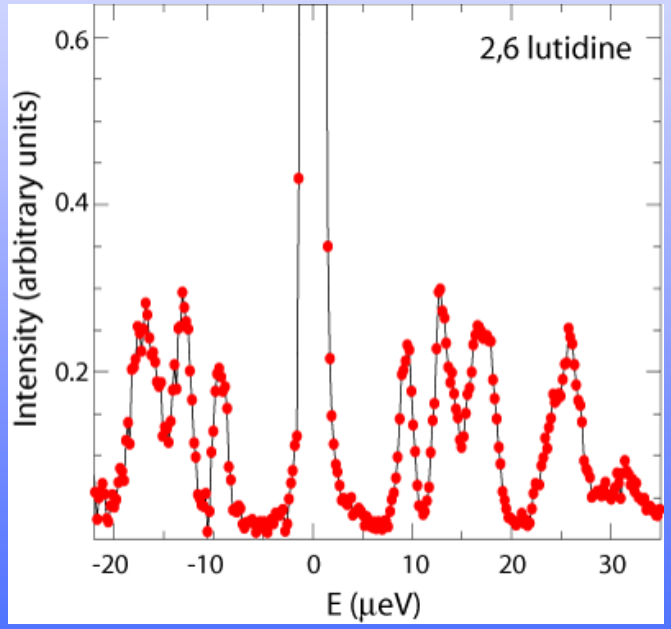
Why HFBS?

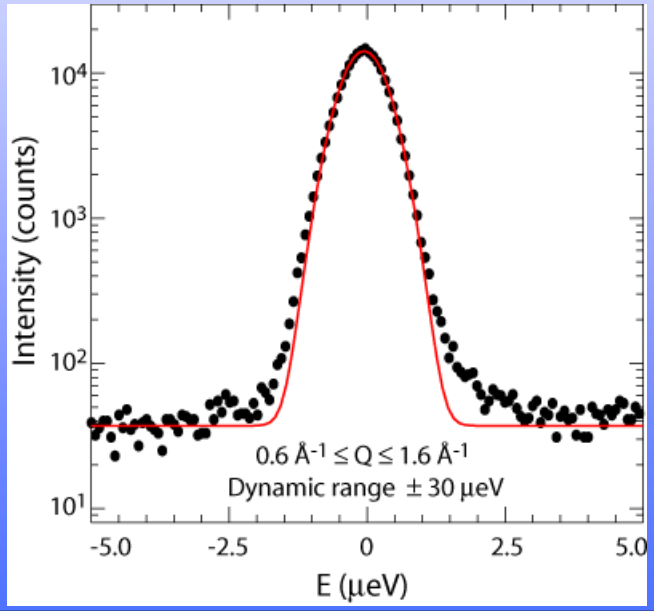
i) PST chopper increases effective flux at sample almost 4 times!

ii) BS with high energy resolution.

iii) Cam-based doppler drive is able to extend the dynamic window to ±36 μeV.







almost Gaussian

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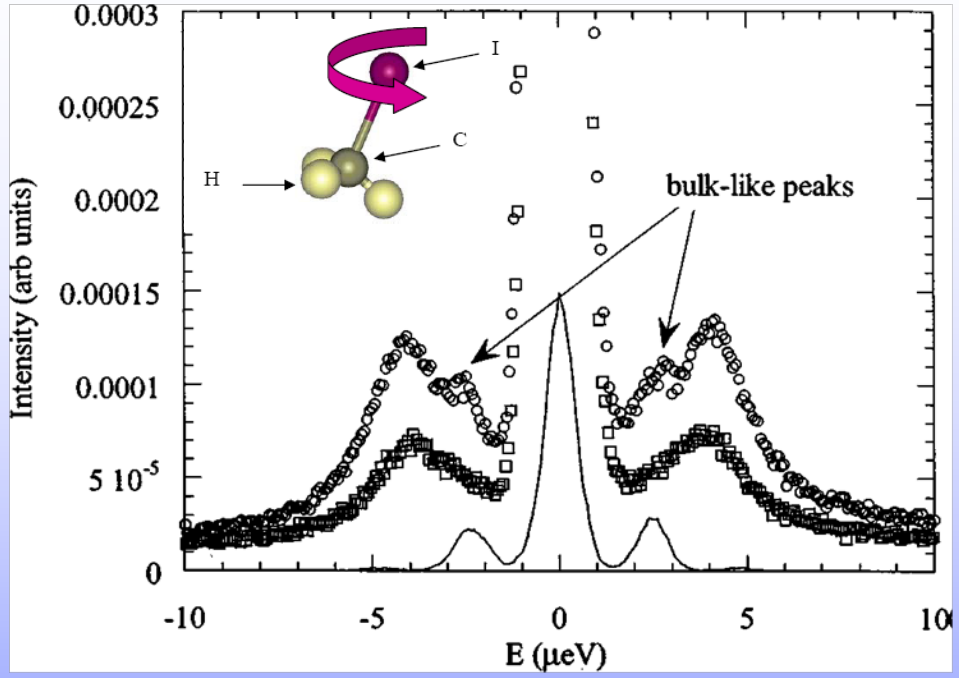
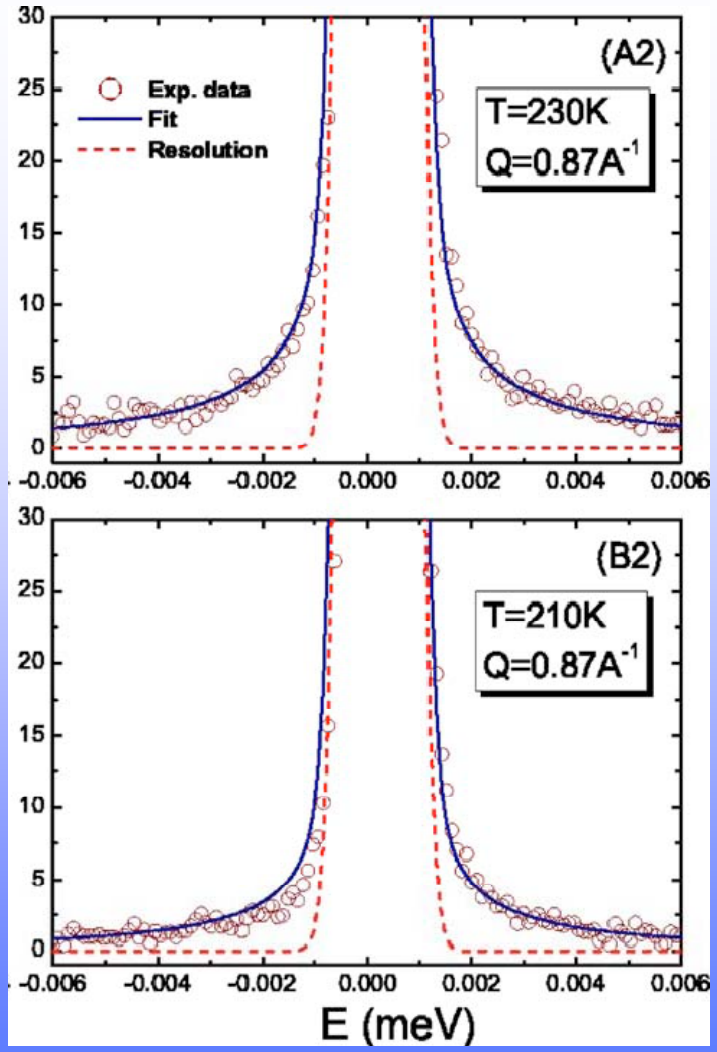
Some Examples: Why HFBS?



Confined methyl iodide

Water in hydrated DNA



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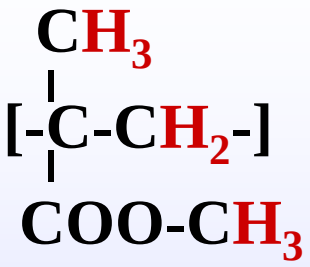
Choosing a right sample

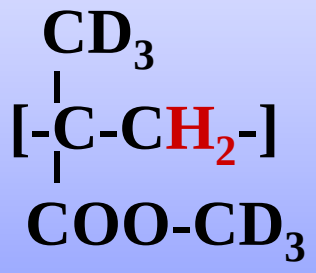
A polymer example: poly(methyl methacrylate) **PMMA:**



>95%

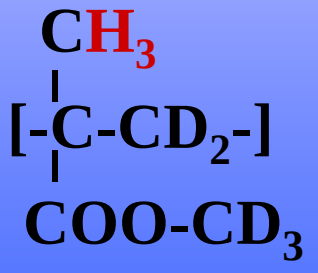
Self motion of PMMA: ⇨ *have it hydrogenated*

Chain motions of PMMA: ⇨ *partially deuterated*



>70%

α-methyl group motions of PMMA: ⇨



>80%

Preparing your sample

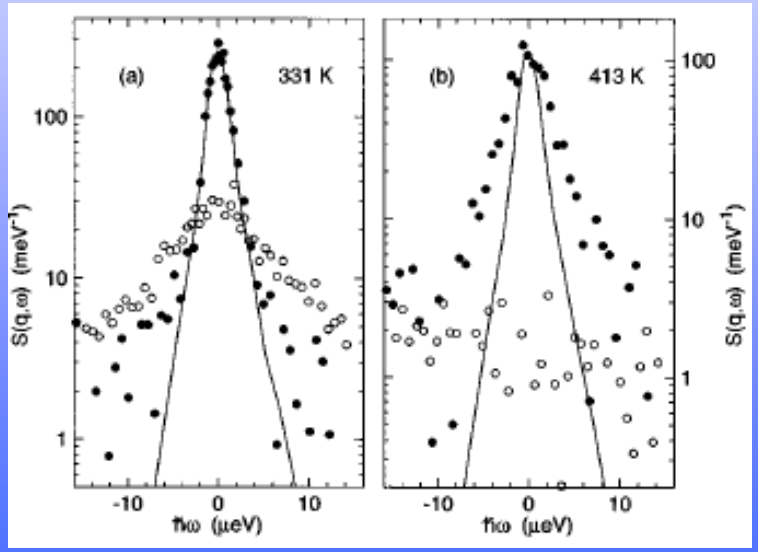
Scattering depends on your sample!

$$T = \exp(-\mu t)$$

where

$$\mu = \rho \times \frac{1}{M} \times N_A \times \sigma_T$$

Transmission T through the sample depends on thickness and total scattering cross section of your sample.

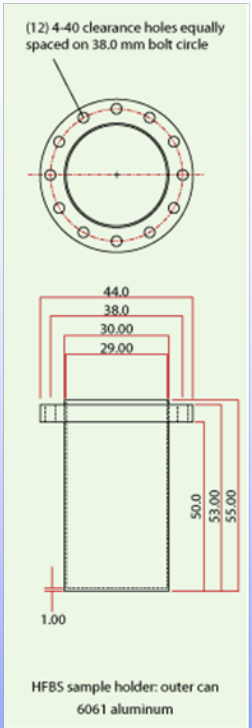


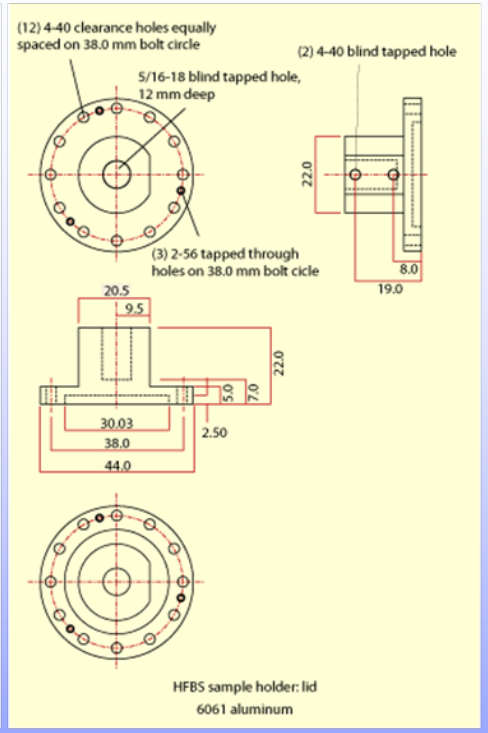
Choose correct thickness:

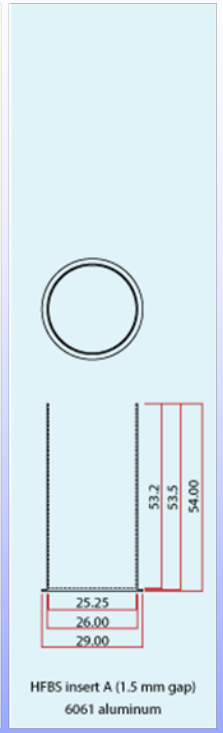
Avoid multiple scattering within the sample.

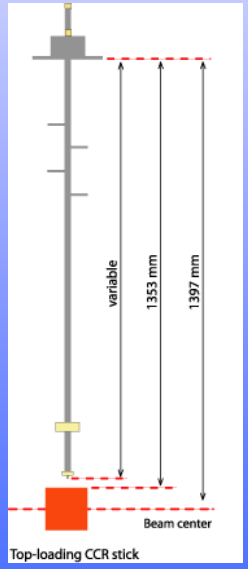
Choose transmission to be about 90%!

Sample environment and cells









Depending on your *T* range, choose
i) correct sample equipment
ii) correct sample cell
iii) correct sealing agent

use neutron-transparent aluminum to fill the extra space.

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Data Reduction

Incoming neutron flux dependence: How to remove it?

Monitor normalization

The factors to be considered to compare theoretical S(Q,ω) with experimental one:

The neutrons counted in a detector at a given solid angle will depend on:

i) Double differential cross section → Sample dependent
ii) Incoming neutron flux
iii) Sample and beam size
iv) Detector efficiency
v) Analyzer efficiency
vi) Analyzer area seen by detectors → Vanadium standard
vii) Scattering from the instrument
viii) Scattering from the empty can → Empty cell

Data Reduction

How to convert measured S(Q,ω) into absolute units?

Incoherent scatterer

Vanadium standard

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DAVE
Data Analysis and Visualization Environment

For complete data reduction:

i) You have to collect sample data
ii) You have to collect vanadium data for normalization, detectors efficiency etc.
iii) You have to collect background data with empty sample can.
iv) You have to collect data for instrumental resolution.

$$\tilde{S}(Q,\omega) = S(Q,\omega) \otimes R(Q,\omega)$$

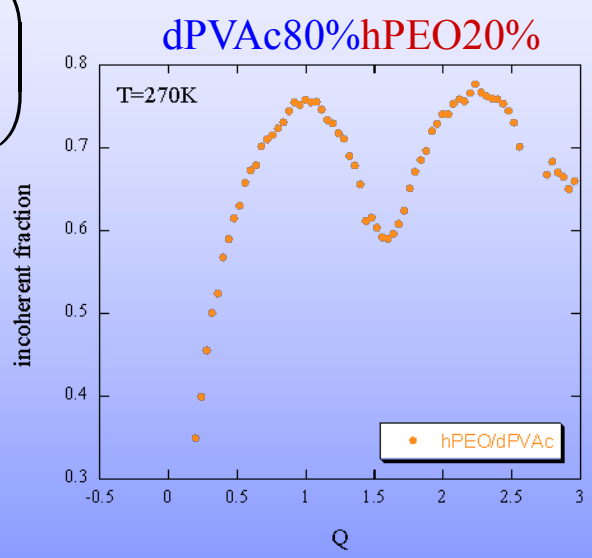
measured — true — resolution

Finally getting S(Q,ω) the one you want!

Maybe not!

Backscattering measures total intensity:

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{k_f}{k_i}\left(\frac{\sigma_{coh}}{4\pi} S_{coh}(Q,\omega) + \frac{\sigma_{inc}}{4\pi} S_{inc}(Q,\omega)\right)$$



Good news is:

If sample is hydrogenated then the coherent part can be ignored.

If sample has non-negligible coherent scattering, then one can make use of neutron diffraction with polarization analysis and separate the two contributions!!

Let's start a backscattering experiment

From where? → Fixed window scans

Make the monochromator and analyzers to reflect neutrons with fixed wave vectors k_i and k_f:

→ Doppler frequency = 0

$$\Delta E = \frac{\hbar^2}{2m}\left(k_f^2 - k_i^2\right)$$

Time scale?

1THz ~ 4.136meV

HWHM for BS: 0.4μeV

time scale ~ 10ns

Example of elastic scans for PVME

Elastic scans scan the motions that are slower than instrumental resolution.

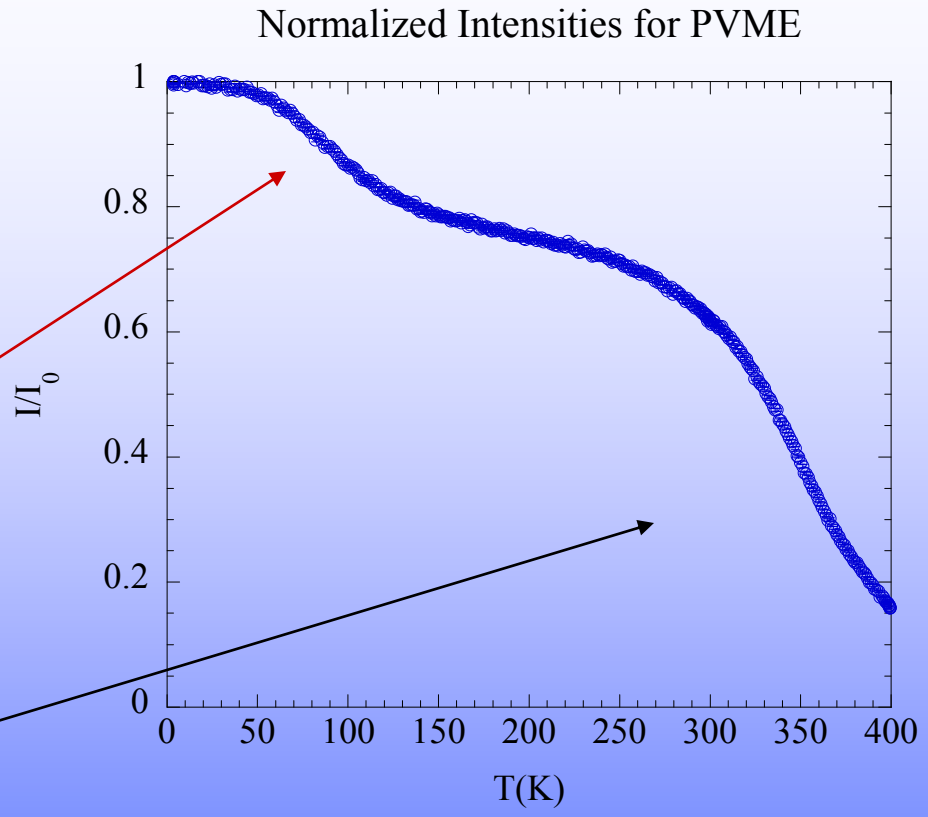
The two step relaxation equivalent of MCT can be seen!

Faster step –

methyl group rotations

Slower Step –

α-relaxation of PVME



$$-[CH(CH_3)-CH_2-]_n$$

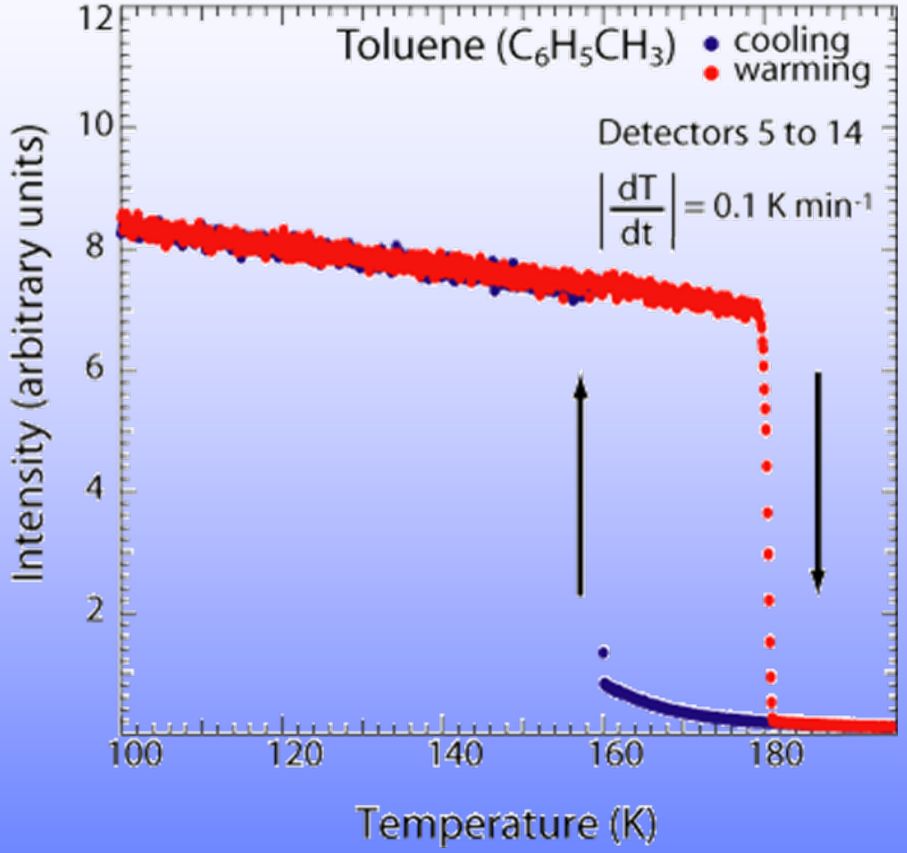
$$O-CH_3$$

Some more examples:

FWS can also be used to study phase diagram of the system.

Sharp transitions ⇨ 1st order

large hysteresis between cooling and heating is indicative of undercooling!



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Backscattering Spectroscopy

Need a dynamic window? ⇒ Turn ON the Doppler!

$$\frac{\delta E_D}{E_i} \approx 2\frac{v_D}{v_i}$$

Velocity of monochromator

Accessible dynamic range for HFBS

Doppler frequency	dynamic range
15Hz	±11μeV
24Hz	±17μeV
50Hz	±35μeV

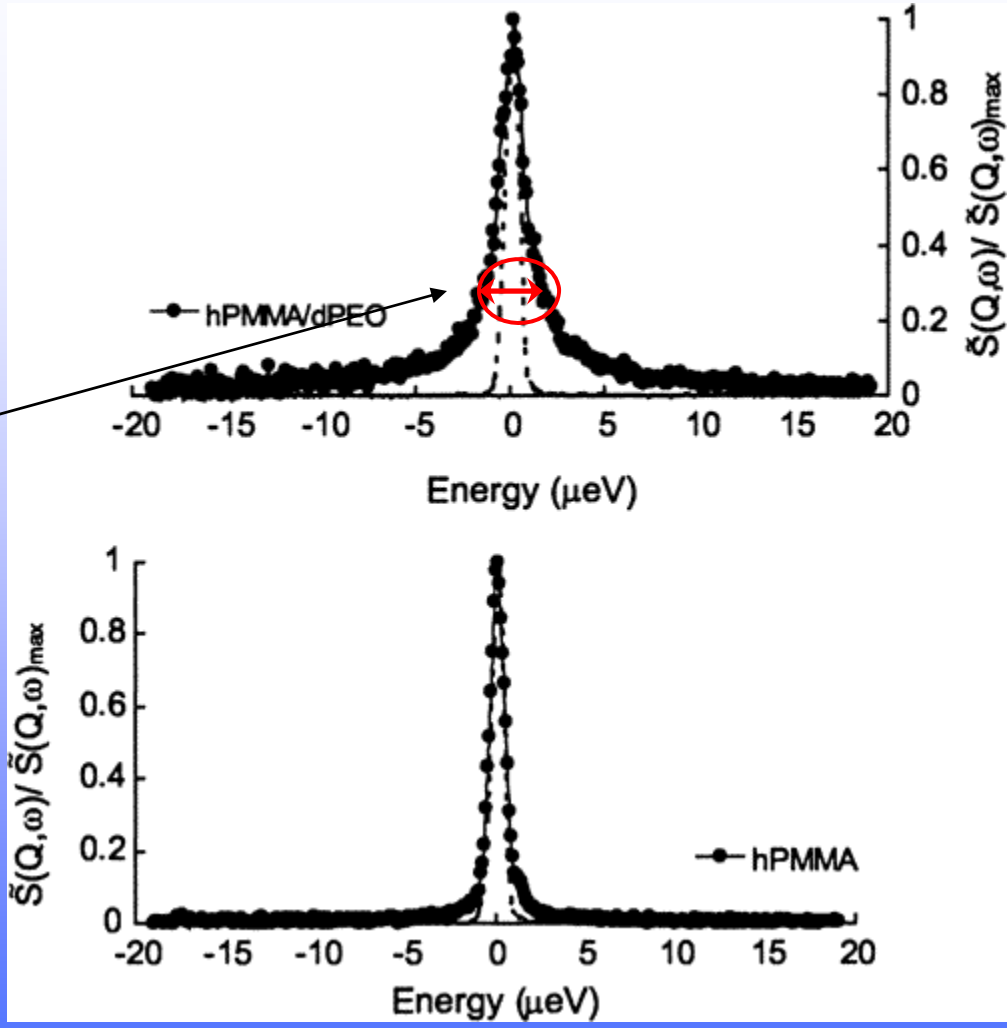
maximum v_D will determine the maximum energy transfer

Some Examples

PMMA and PMMA with PEO:

PMMA: T_g=400K
PEO: T_g=220K

Relaxation time τ is inversely proportional to the width w.r.t. instrumental resolution!



PEO acts as a plasticizer for high T_g component.

Acknowledgments

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Dan Neumann and Yamali Hernandez