Disordered Materials: Glass physics

> 2.7. Introduction, liquids, glasses

> 4.7. Scattering off disordered matter:
> static, elastic and dynamics structure factors

> 9.7. Static structures:
> X-ray scattering, EXAFS, (neutrons), data interpretation

> **11.7. Dynamic structures and the glass transition**

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Methoden moderner Röntgenphysik II
July 2013

DESY

Dynamic structure factor:

$$S(q,\omega) = \frac{1}{N} \sum e^{-iq(R-R')} \int \frac{dt}{2\pi} \left\langle e^{iqu(R',0)} e^{-iqu(R,t)} \right\rangle$$

FT of the density-density correlation function

Energy resolved inelastic scattering !

Time correlation: speckle spectroscopy

$$S(q,t) = \frac{1}{N}\sum e^{-iq(R-R')}\left\langle e^{iqu(R',0)}e^{-iqu(R,t)}\right\rangle$$

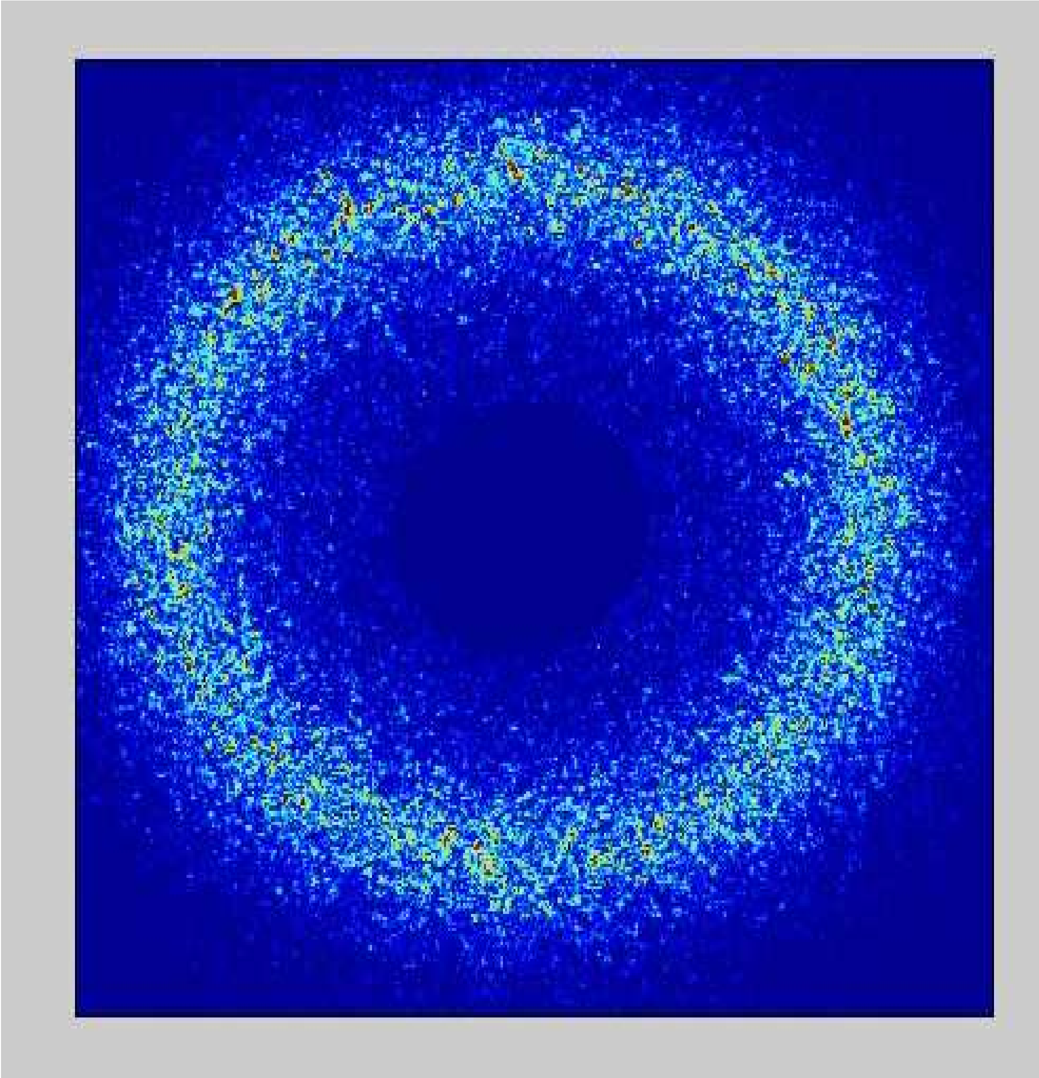
However, we cannot measure the phases

$$S(q,t) \approx \left\langle I(q,0) I(q,t)\right\rangle$$

This is – regardless of normalization - the time correlation function (see lecture 10)

$$g_2(Q,t) = \left\langle I(Q,0) I(Q,t)\right\rangle / < I(Q) >^2$$

Dielectric spectroscopy



Colliodal glass,

70 nm spheres

2000 frames

4 GB

400 s exposure

Courtesy Ch. Gutt

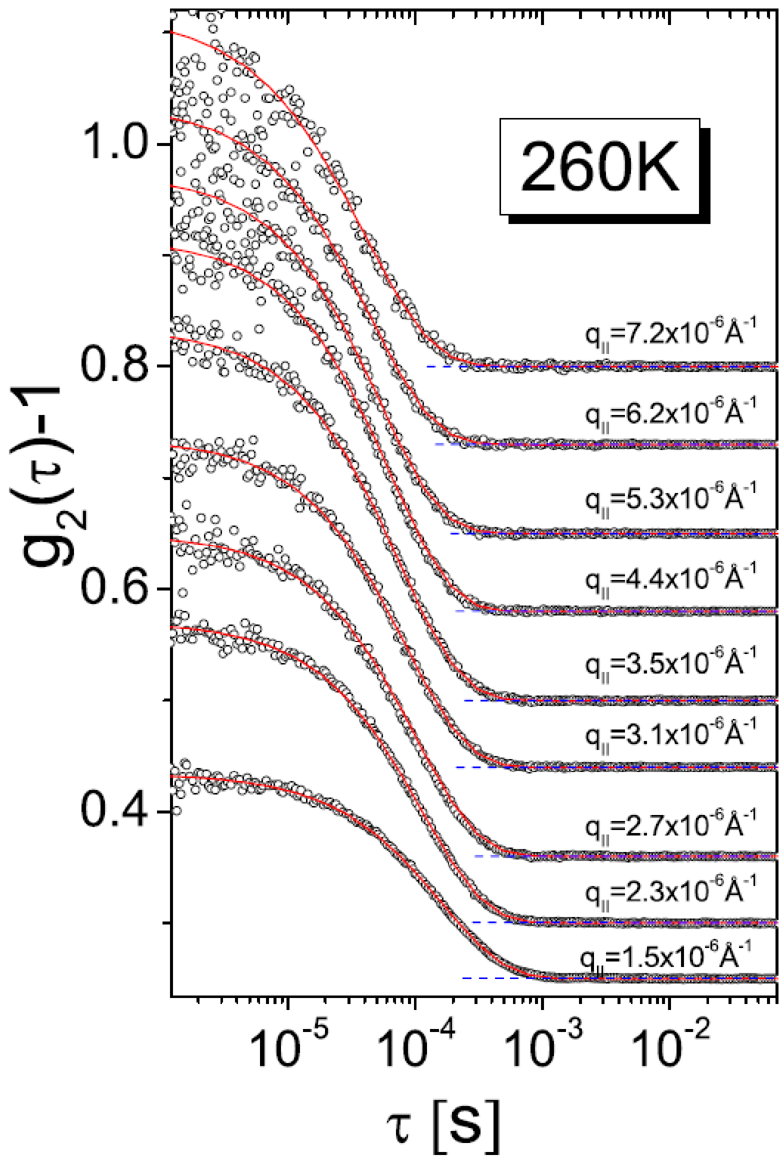


Figure 6.19: Intensity autocorrelation functions $g_2(\tau, q_x)$ recorded on the surface of dibutyl phthalate at 260 K. The origin of the correlation functions have been shifted for clarity and marked with the dashed blue line.

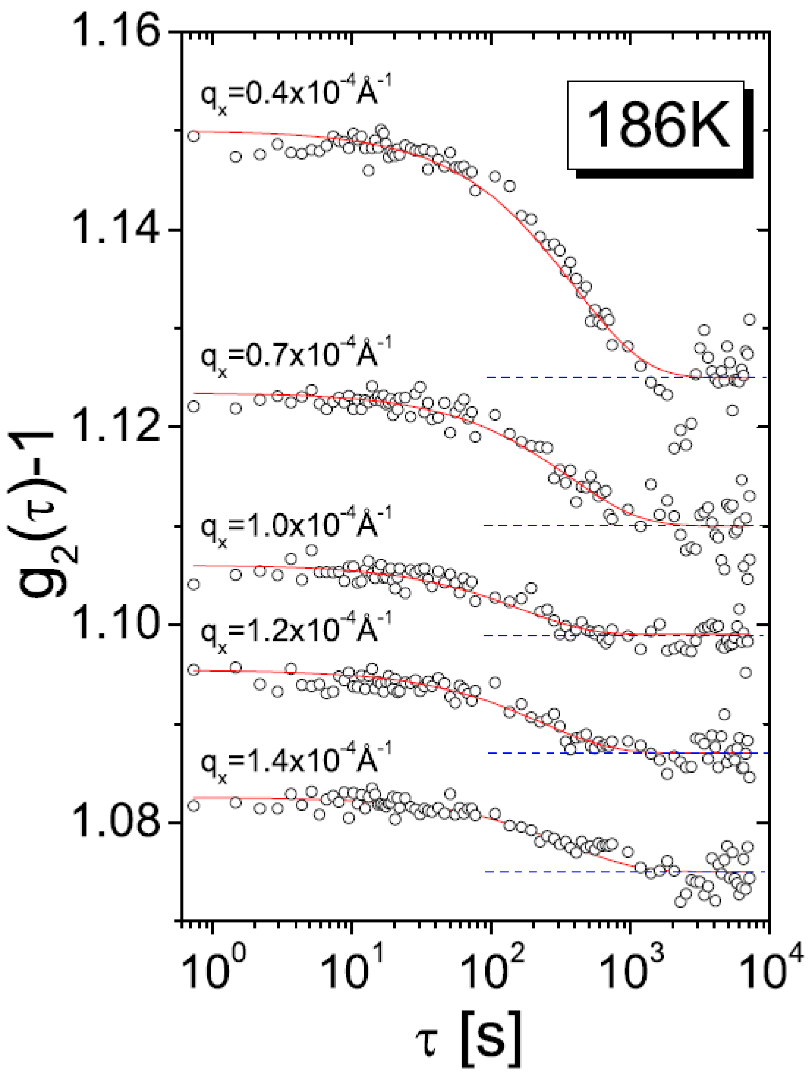
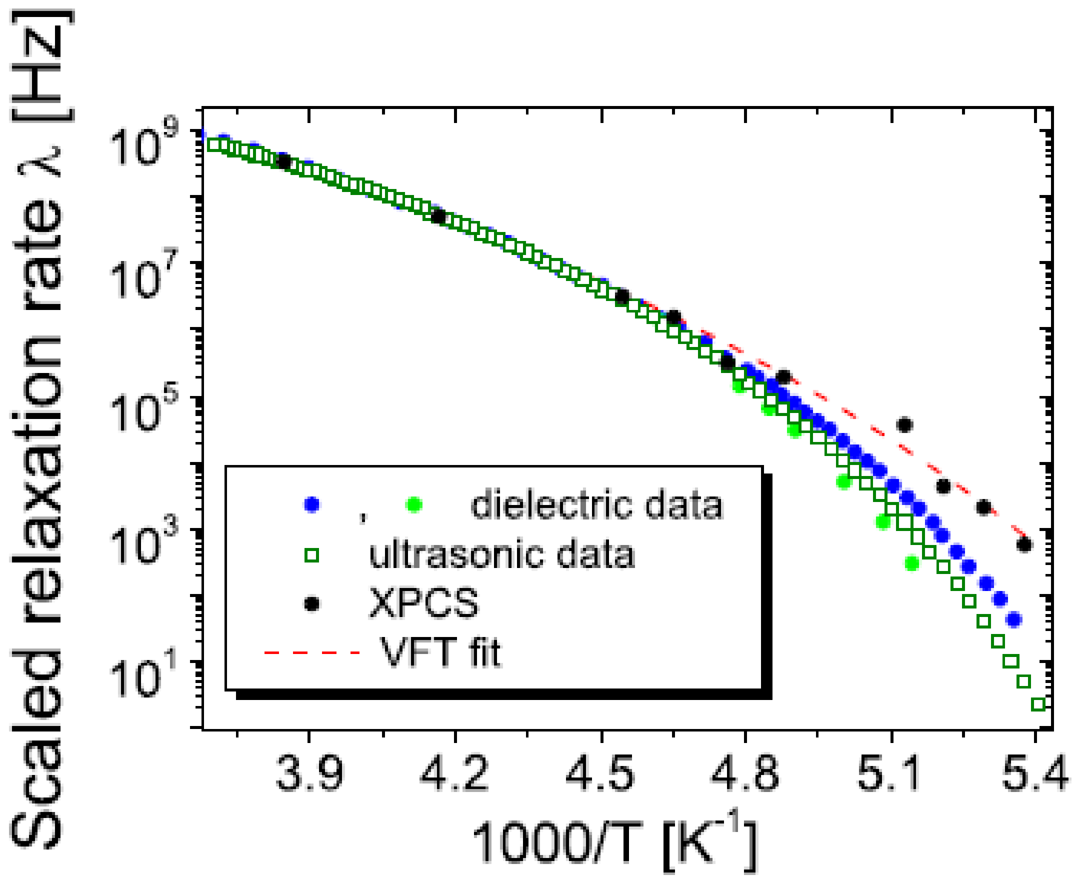


Figure 9.9: Selected intensity autocorrelation functions $g_2(\tau, q_x)$ recorded on the surface of dibutyl phthalate at 186 K. The origin of the correlation functions have been shifted for clarity and marked with the dashed blue line.

$g_2(Q,t) = 1 + ß(Q)\,|f(Q,t)|^2$ and $f(Q,t) = \exp(-\Gamma t) = \exp(-t/\tau)$

In glassy systems $f(Q,t) = \exp(-t/\tau)^{\beta} = \exp(-t^{*}\lambda)^{\beta}$



Dielectric spectroscopy

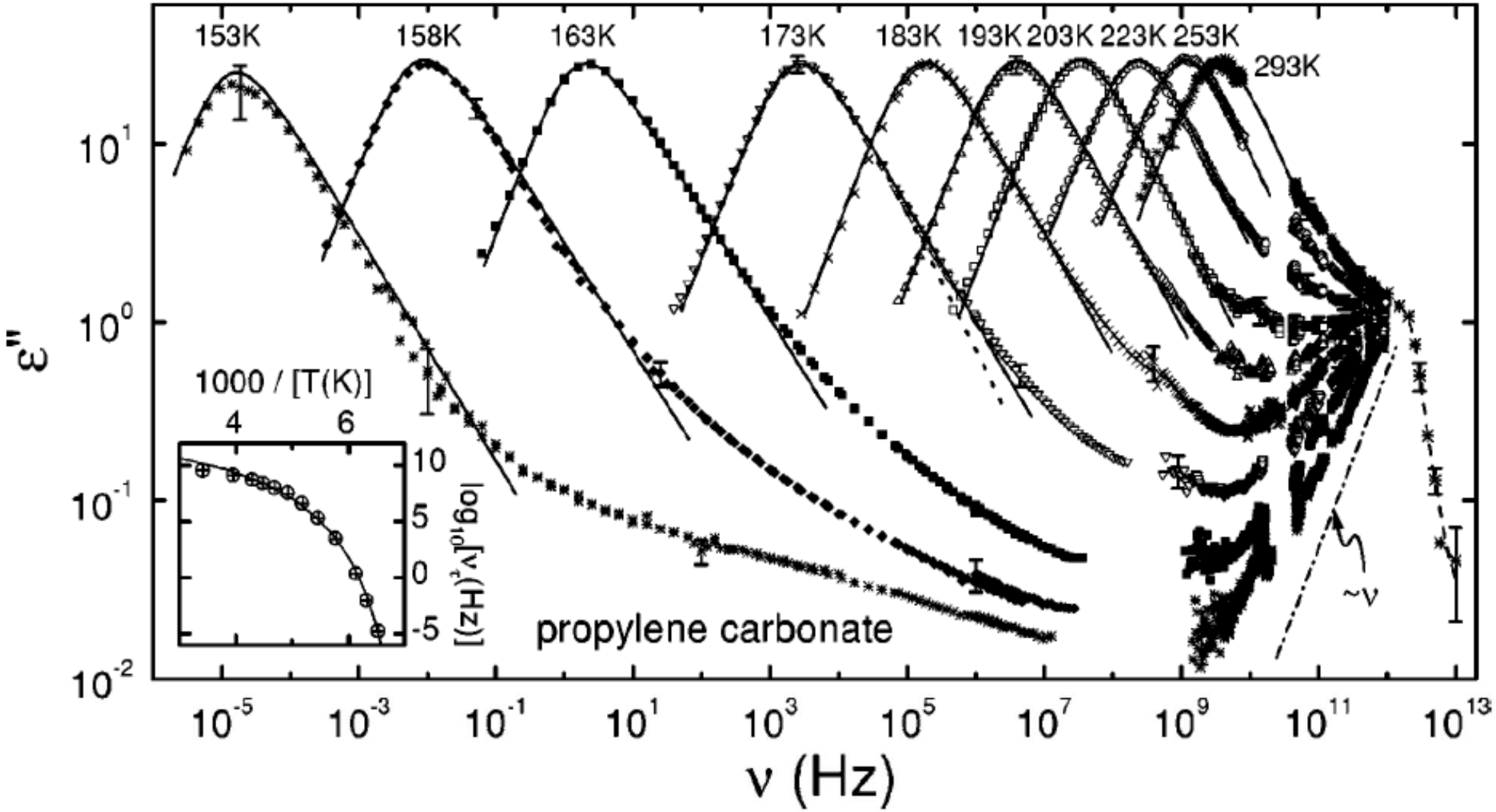


FIG. 2. Frequency dependence of the dielectric loss in propylene carbonate at various temperatures. The solid lines are fits with the CD function, the dotted line is a fit with the Fourier transform of the KWW law, both performed simultaneously on ε'. The dash-dotted line indicates a linear increase. The FIR results have been connected by a dashed line to guide the eye. The inset shows $\nu_\tau = 1/(2\pi\langle\tau\rangle)$ as resulting from the CD (circles) and KWW fits (pluses) in an Arrhenius representation. The line is a fit using the VFT expression, Eq. (1), with $T_{VF} = 132$ K, $D = 6.6$, and $\nu_0 = 3.2 \times 10^{12}$ Hz.

U. Schneider et al. PRE (1999)

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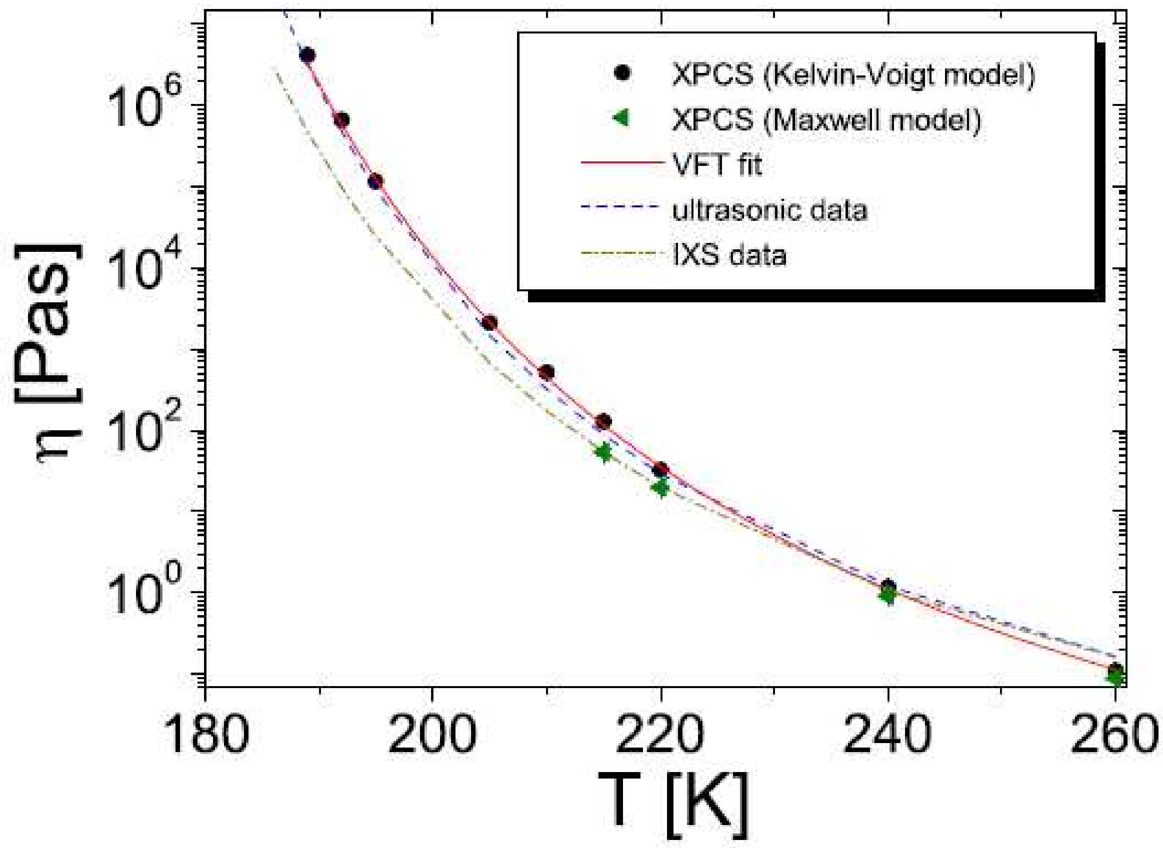

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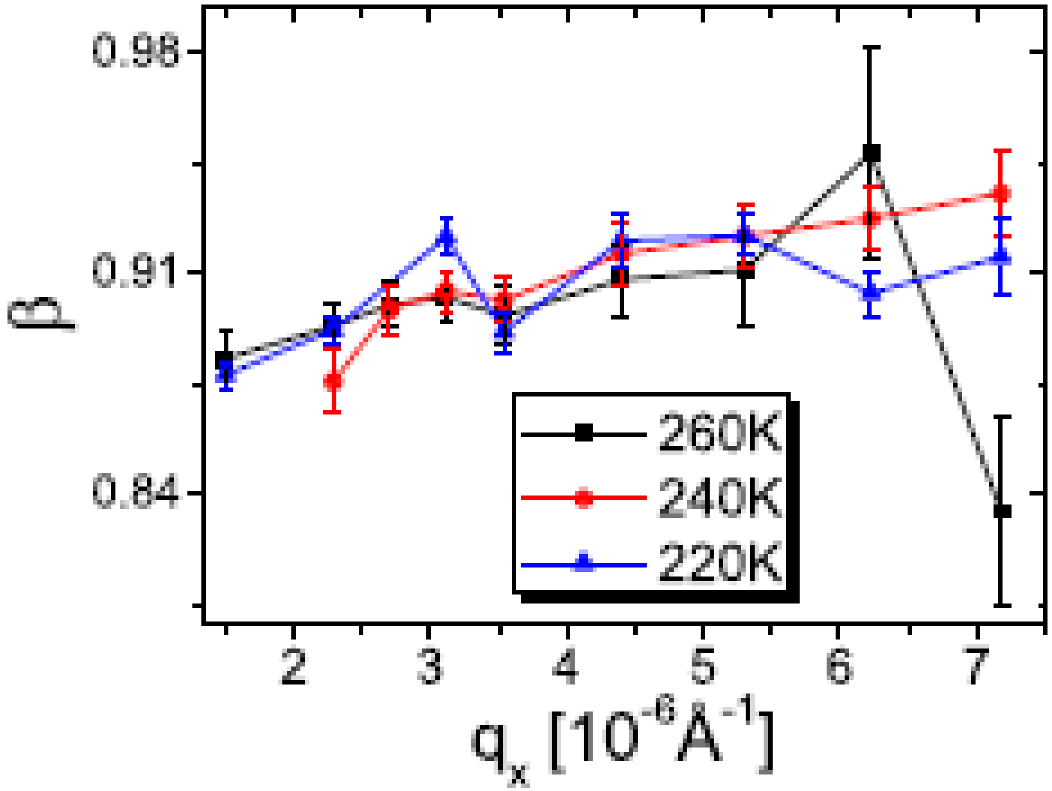


Nuclear resonant scattering

Nuclear resonant scattering

Up to now all lectures treated scattering from electrons

$$r_e = \frac{e^2}{mc^2} = 2.818\ 10^{-15}\ \text{m} = 2.818\ 10^{-5}\ \text{Å}$$

electron : nucleus : 511 keV : 938,280 keV

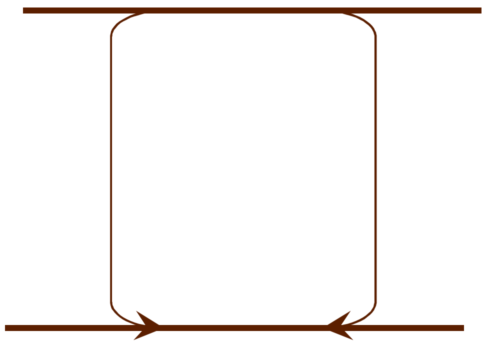
Thompson scattering of nuclei is negligible

0.0005 effect in the amplitude

Resonant scattering

$$f(\omega) = \sum_n \frac{\bar{\lambda}_0 \Gamma_R}{(E_n - E_g) - \hbar\omega - i\Gamma_T/2}$$

$$f(\omega_0) = \frac{\bar{\lambda}_0 \Gamma_R}{\Gamma_T/2}$$



excited state $E_g + \hbar\omega_0$

ground state

holds for (any) resonance

Resonant scattering

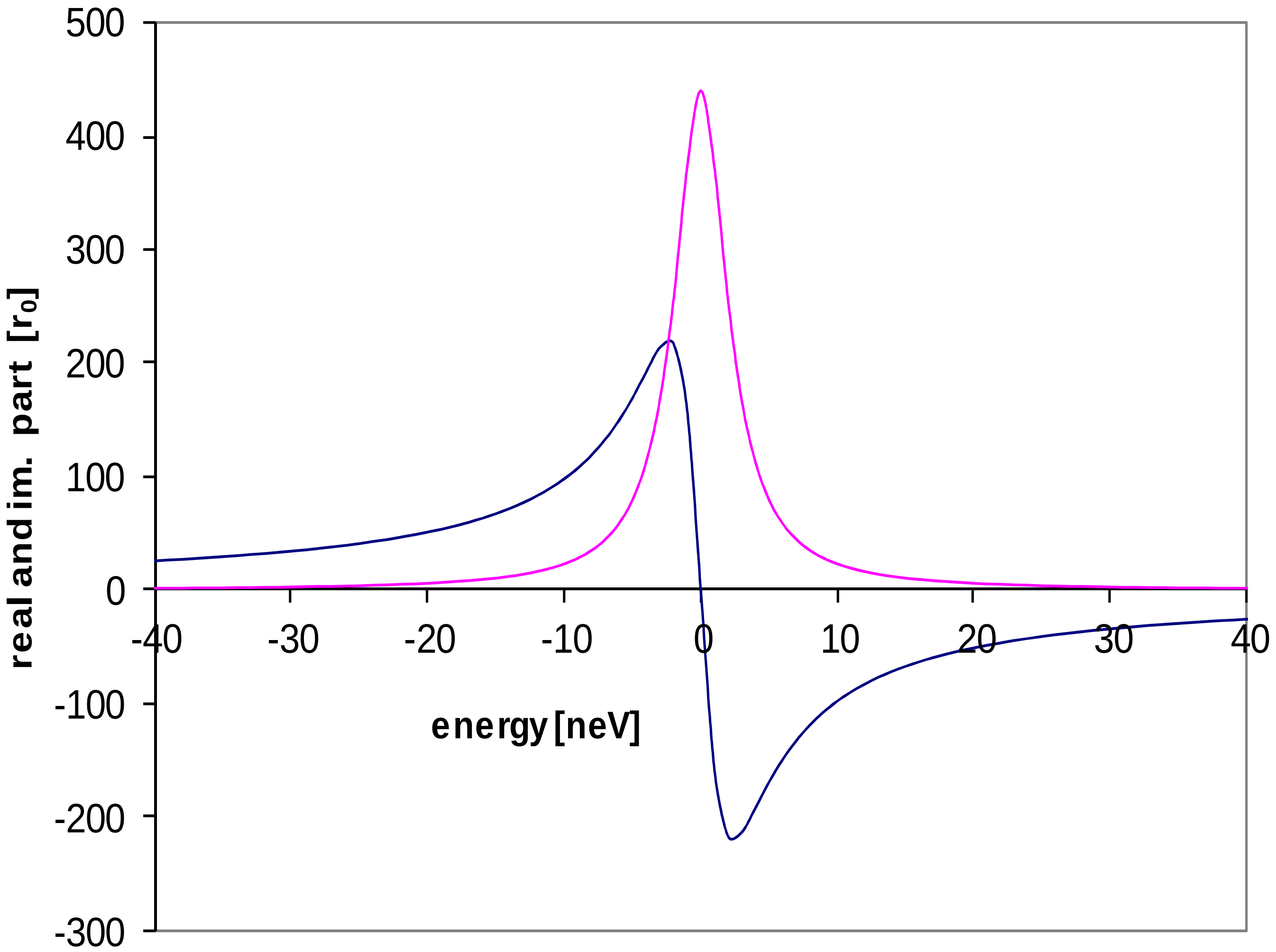
electrons: $$\lambdabar = \frac{\lambda}{2\pi} \cong 0.2\text{Å} \qquad \Gamma_R \approx 0.005\Gamma_T \quad (eV)$$

nuclei: $$\lambdabar = \frac{\lambda}{2\pi} \cong 0.2\text{Å} \qquad \Gamma_R \approx 0.1 - 0.8\Gamma_T \approx neV - \mu eV$$

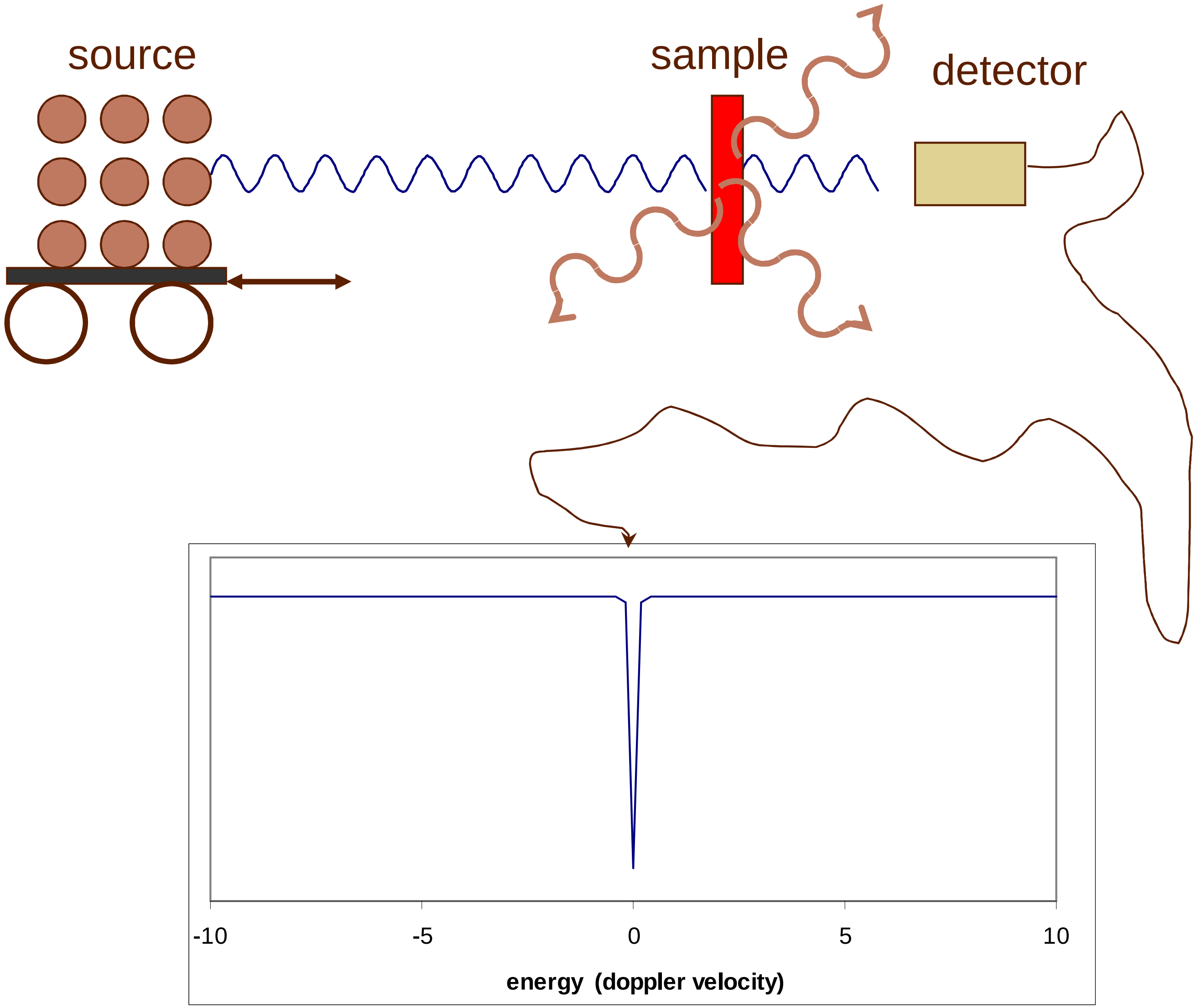
$$\mathrm{Re}(f(\omega_0, {}^{57}Fe)) \approx 440 r_0$$

under resonance conditions the cross-section of nuclei excceeds the scattering from electrons

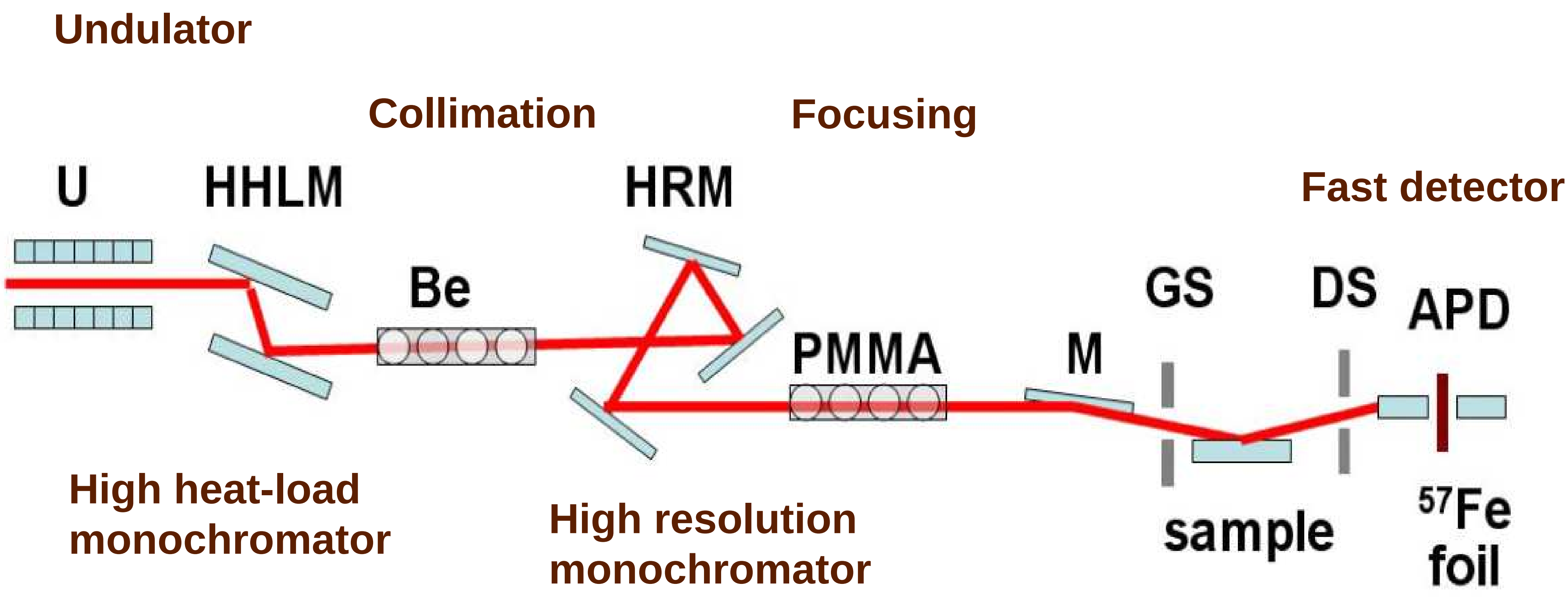
^{57}Fe Resonance



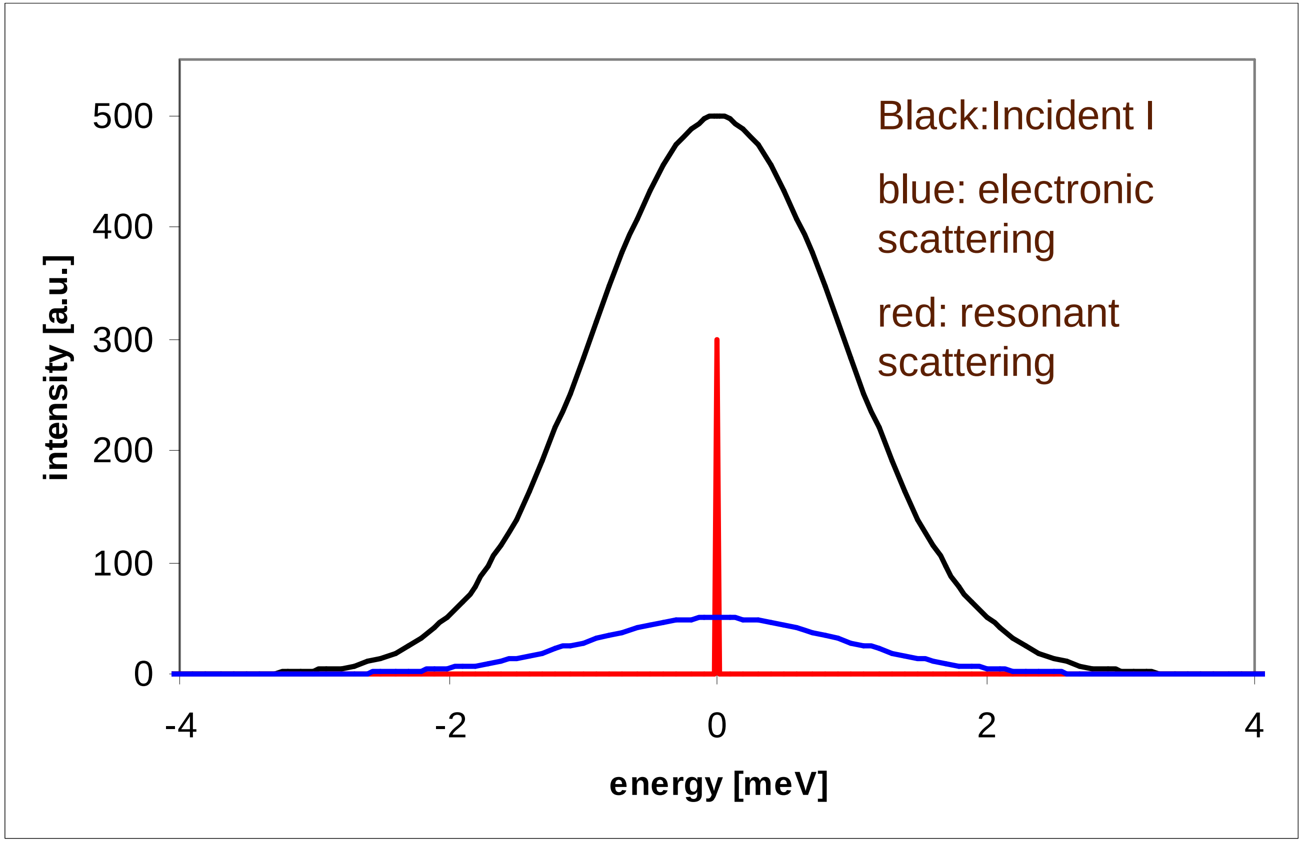
Excursion: The Mössbauer effect



Experimental setup

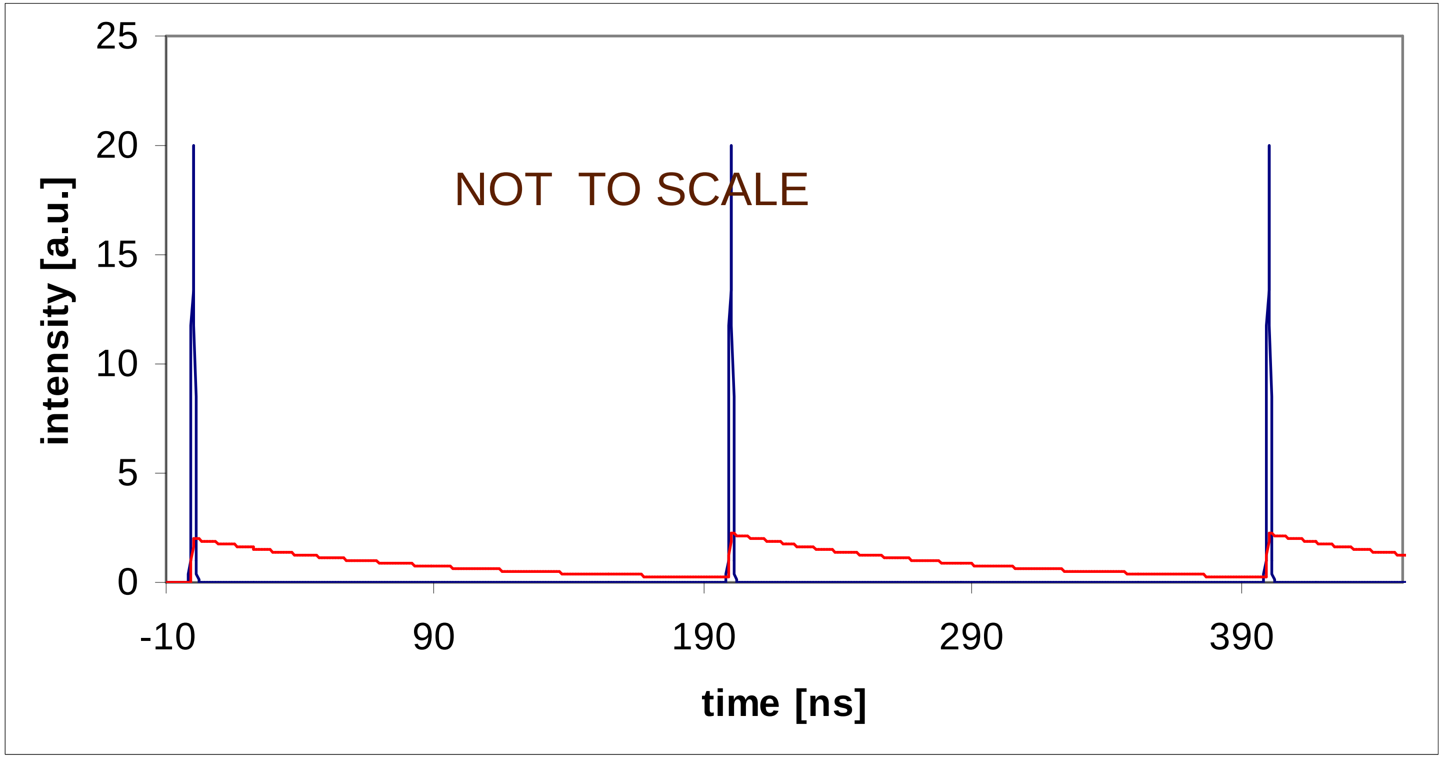


Problem: electronic background



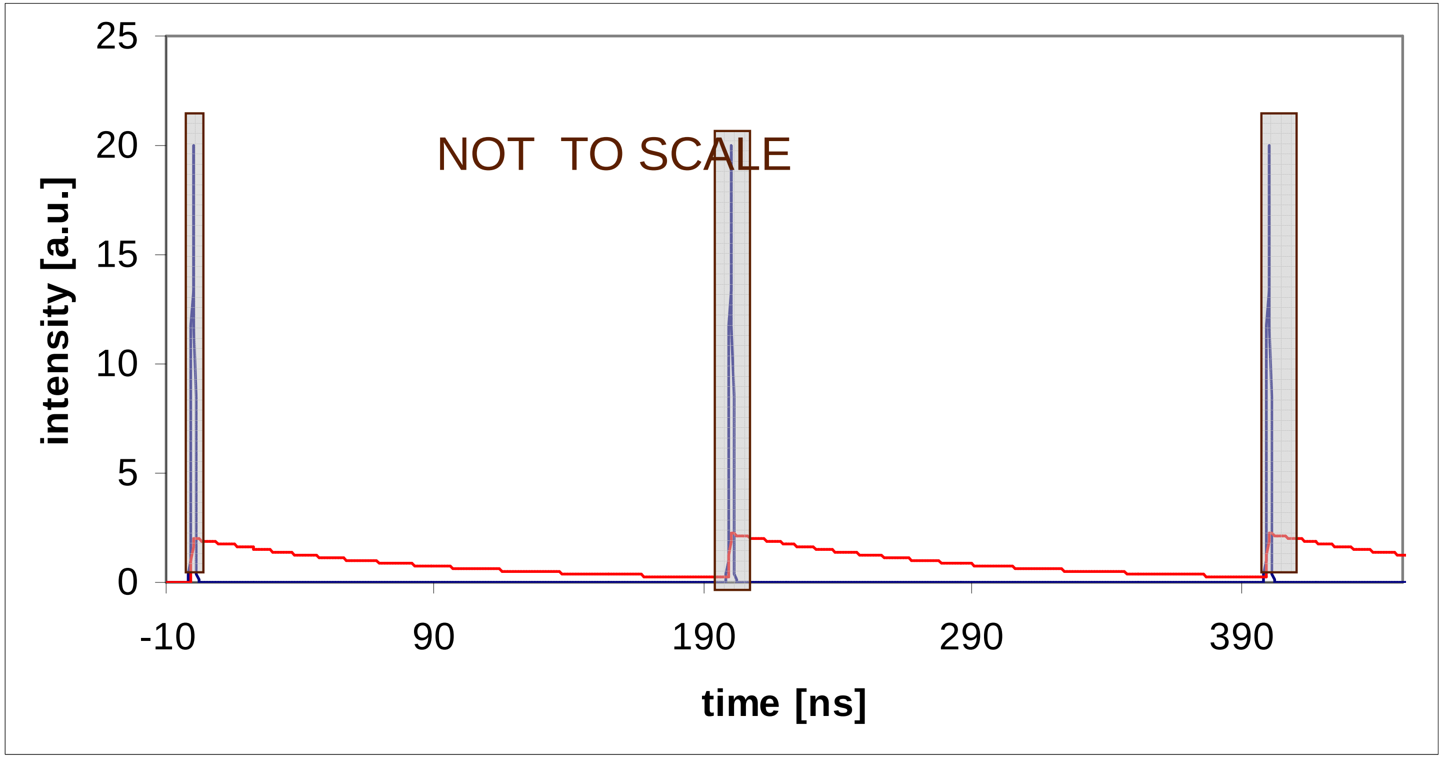
Resonant scattering is strong but limited to extremely narrow bandwidth (neV)

Way out: timing



Due to the narrow bandwidth the response is slow (long life time, Heisenberg)

Way out: timing



Electronic gating (of the detector) takes away the fast scattering off electrons

SO WHAT ???

Is there any advantage in using scattering off the nuclei?

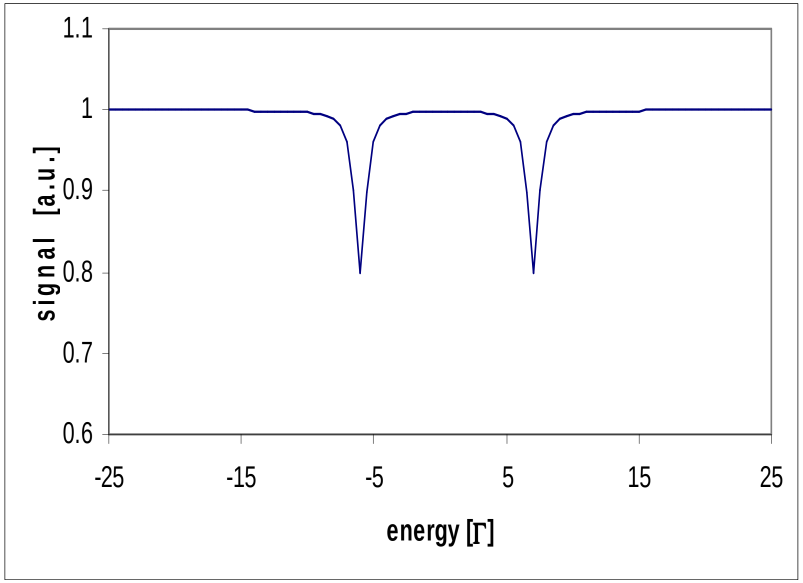
Yes !

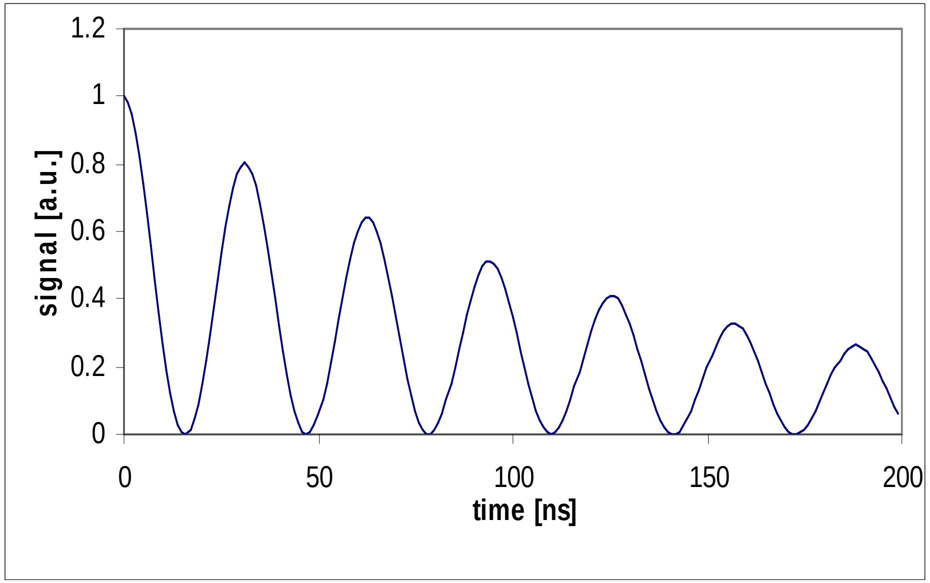
- <u>SR is an ideal source</u>: no line broadening, no back ground (after gating), high brilliance, no radioactive source
- direct observation in time domain
- “white” incident radiation offers the possibility to perform inelastic measurements

Nuclear exciton

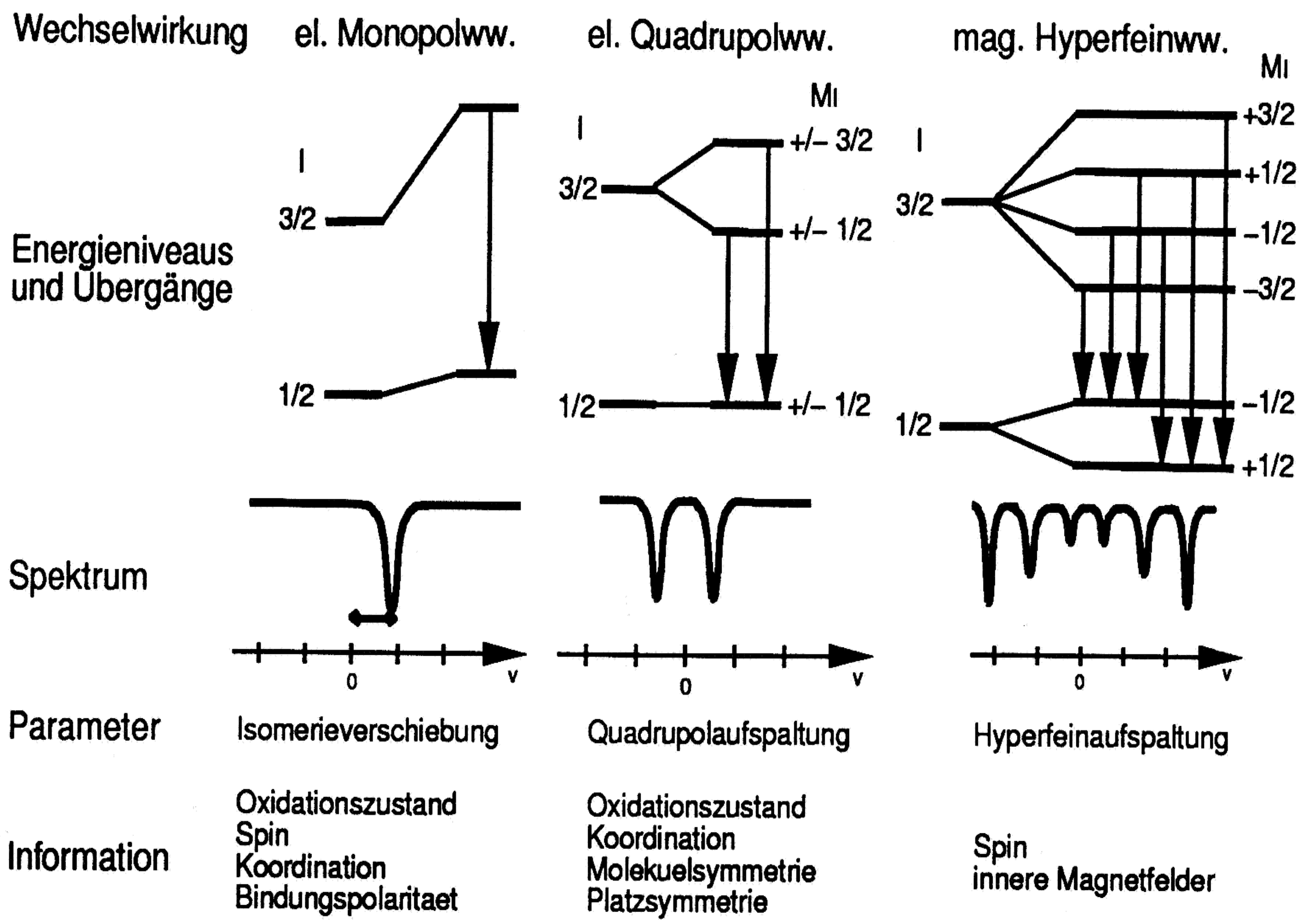
The incident pulse excites all nuclei coherently, no nucleus is distinguished

Thus the response is the sum of all scattering AMPLITUDES

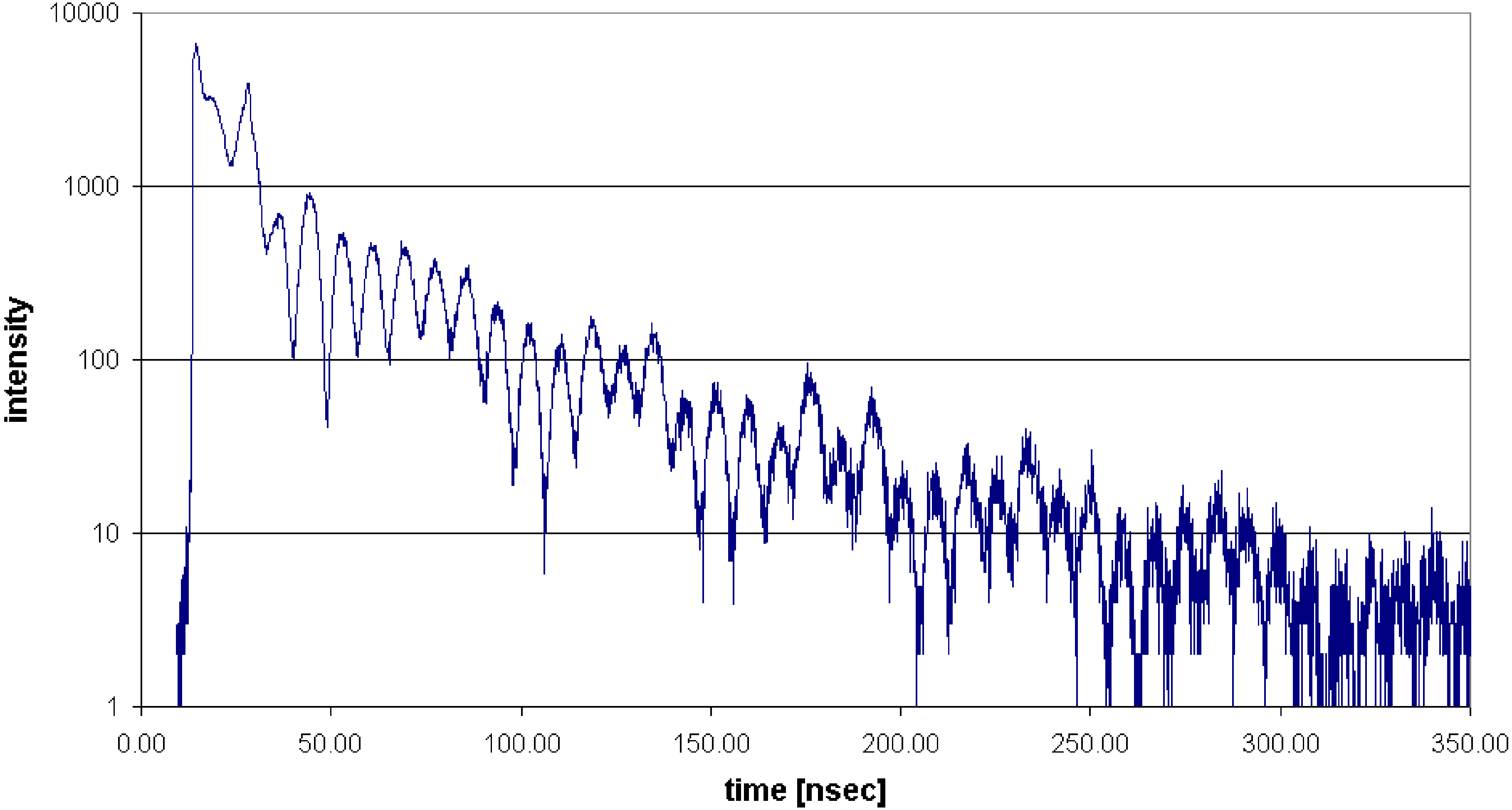




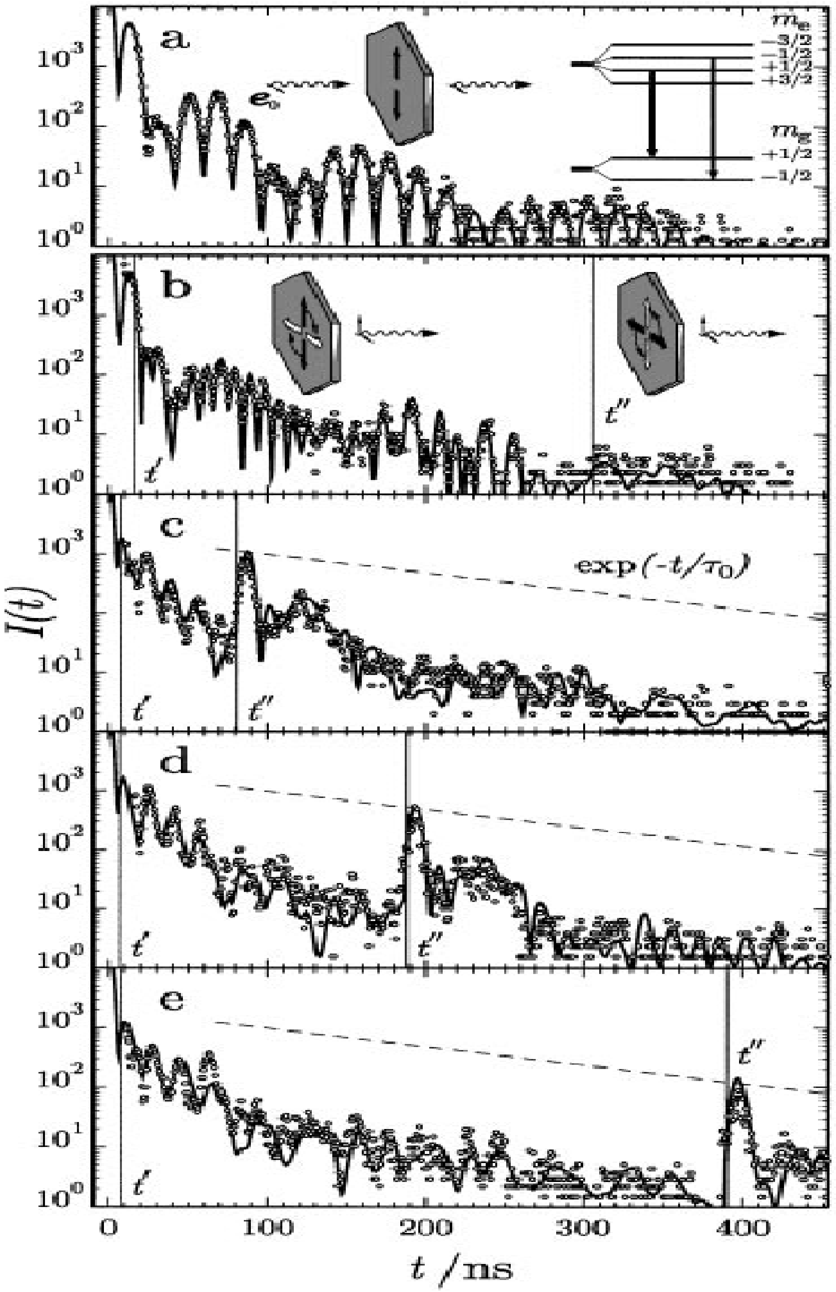
Types of hyperfine interactions



Time spectrum of $FeBO_3$



Magnetic switching



Determine switching angle and time

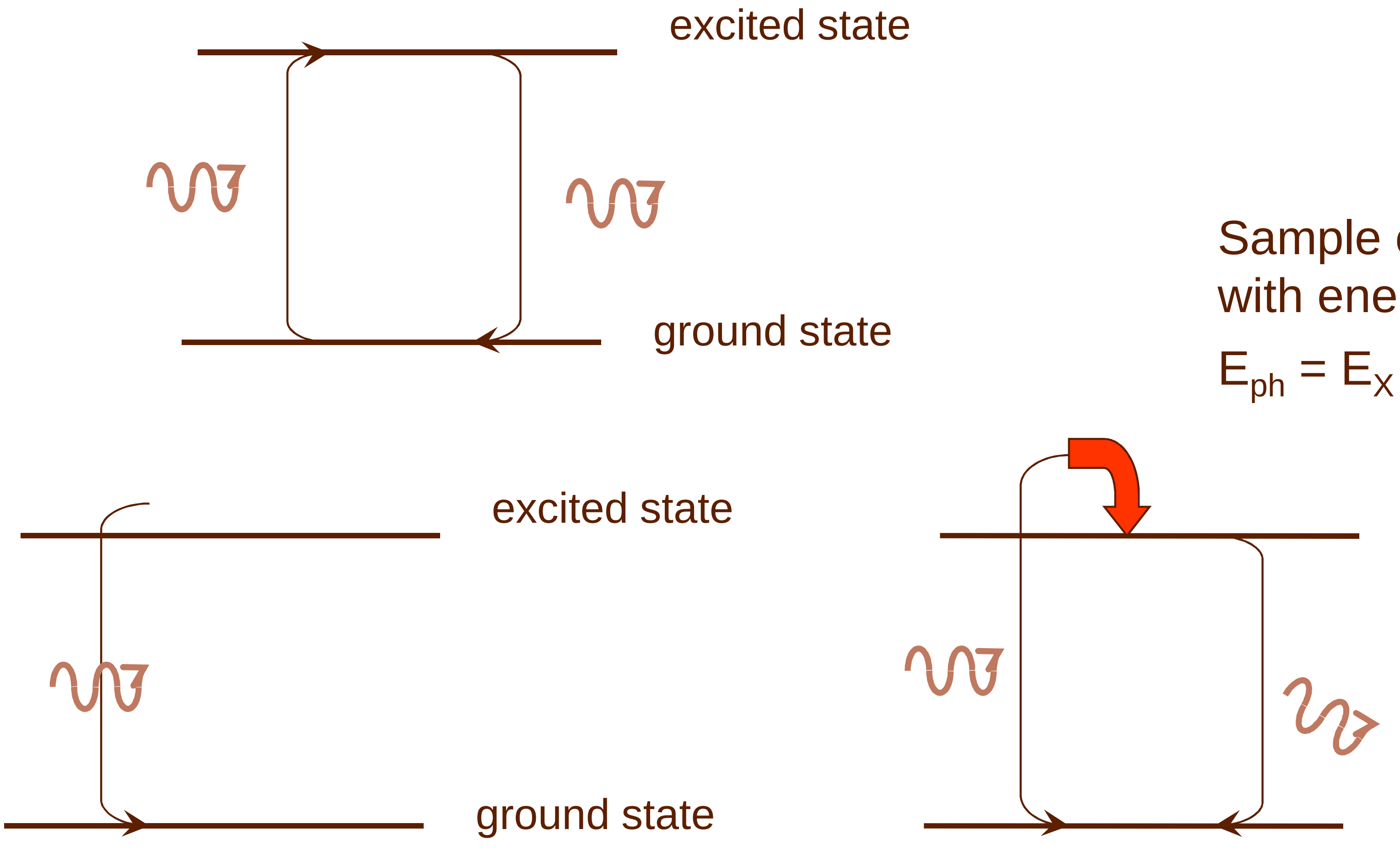
Yu. Shvyd'ko et al. PRL 1996

Resonances suitable for NRS

^{57}Fe	14.412 keV	141 ns	4.7 neV
^{151}Eu	21.5 keV	13.7 ns	48.3 neV
^{161}Dy	25.651 keV	39.2 ns	16.2 neV
^{119}Sn	23.88 keV	25.6 ns	25.8 neV
^{61}Ni	67.41 keV	7.6 ns	87 neV

and several more

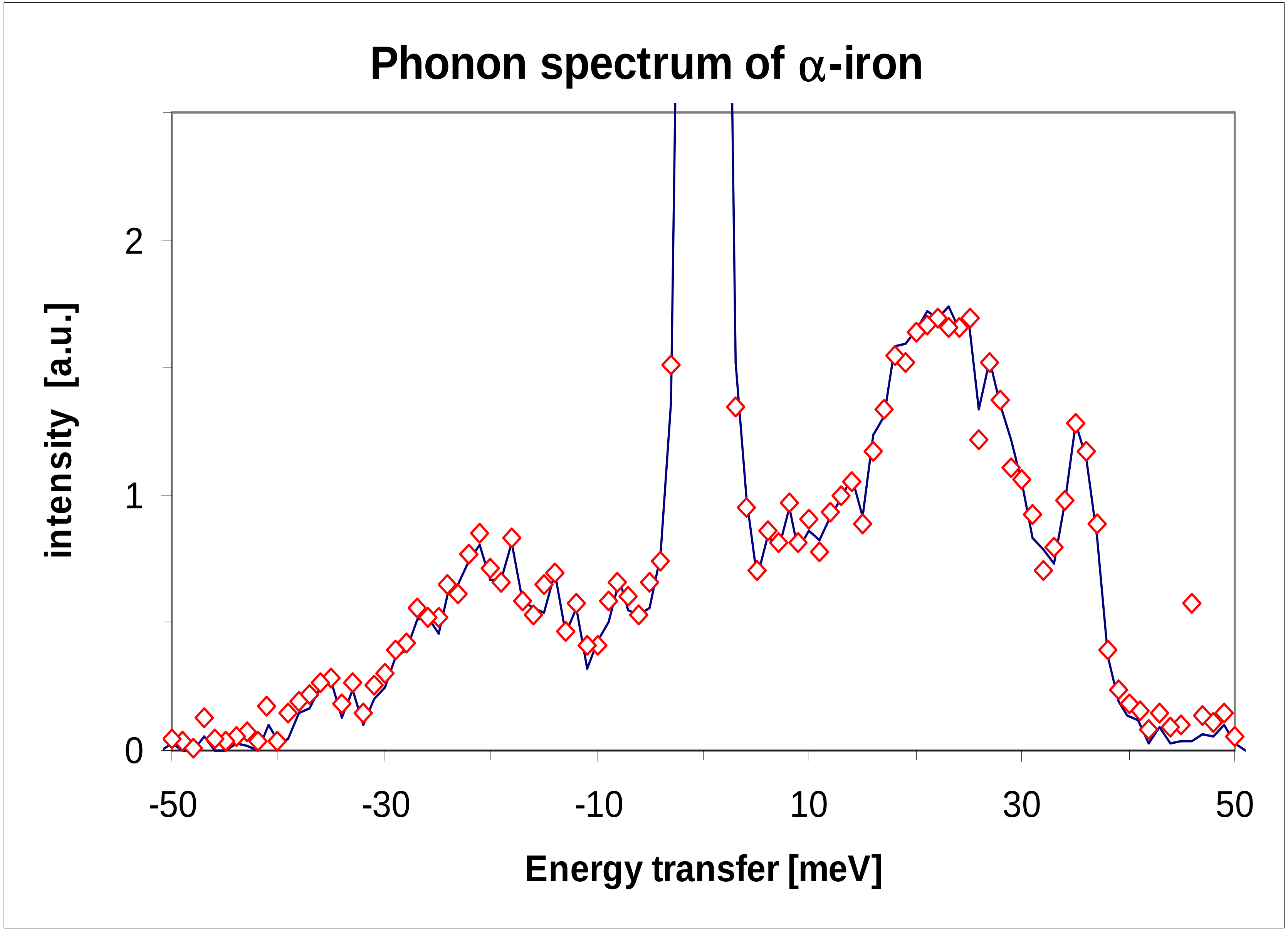
Inelastic scattering



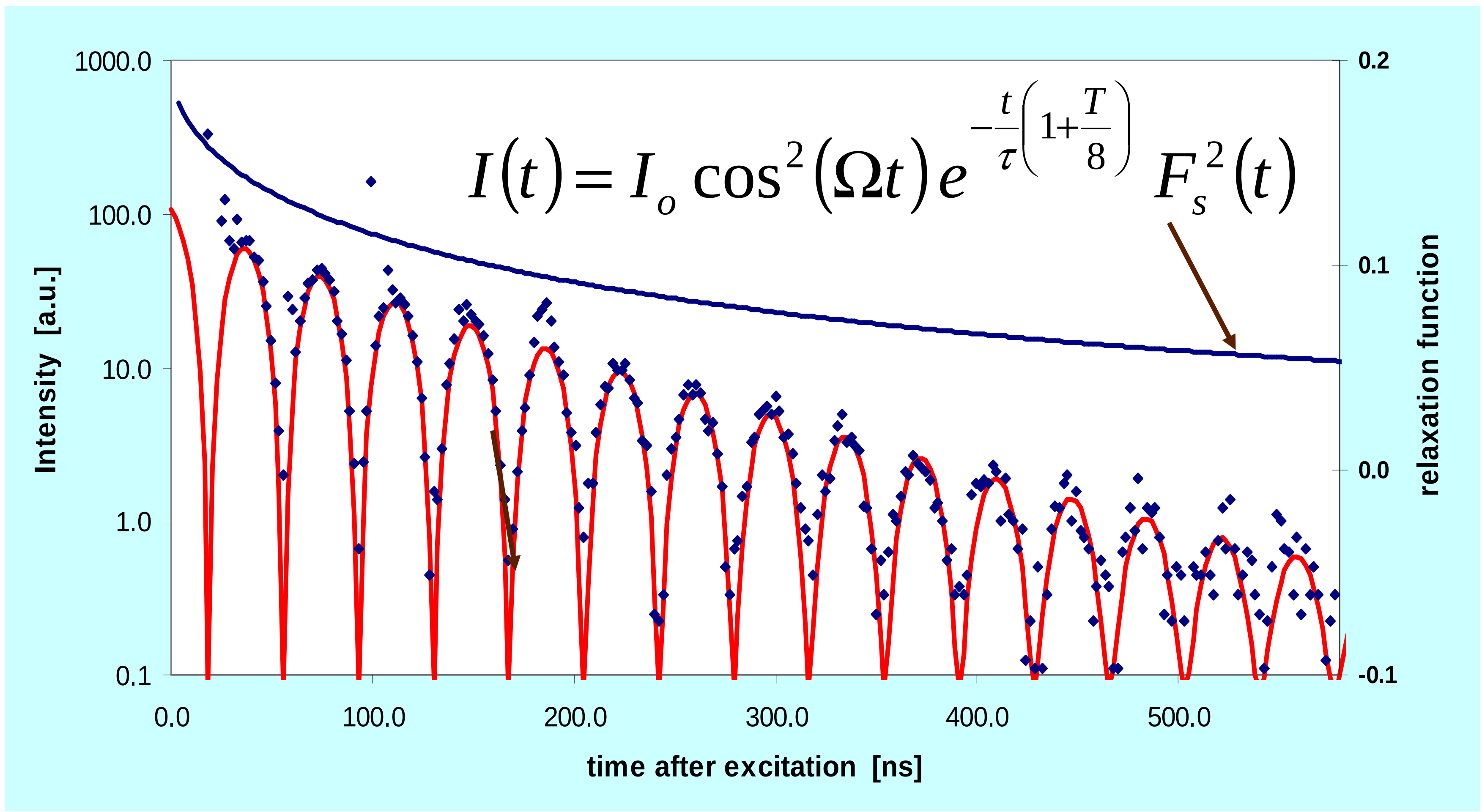
Sample excitation with energy

$E_{ph} = E_X - (E_e - E_g)$

Energies not to scale (keV and meV)


Phonon spectrum of α-iron
intensity [a.u.]
2
1
0
-50
-30
-10
10
30
50
Energy transfer [meV]

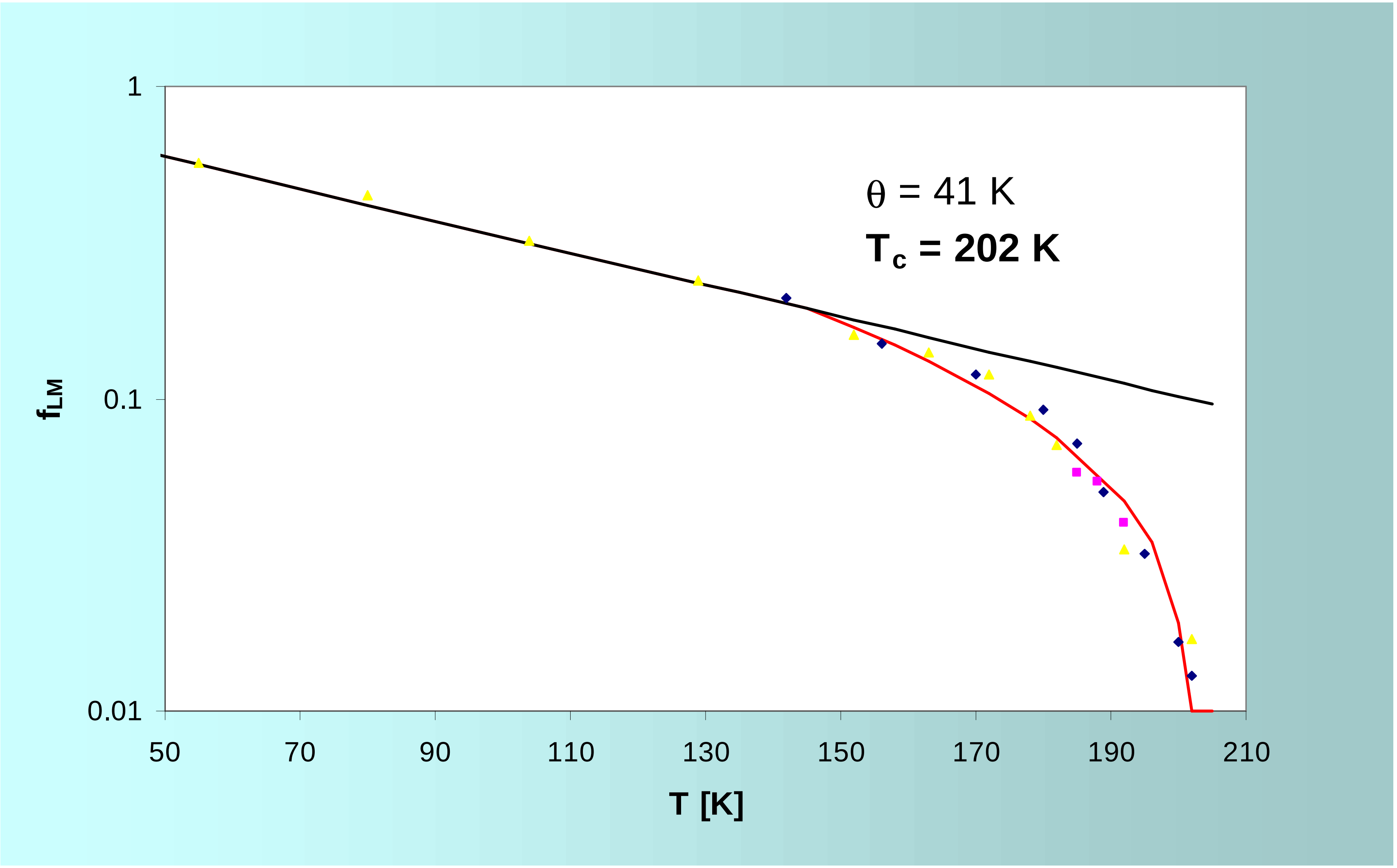
Quasielastic nuclear resonant forward scattering



Butyl phthalate / ferrocene

Exact treatment of QNFS: I. Sergueev, HF,.. PRB 2003

Non ergodicity parameter



Square-root behaviour as predicted by mode-coupling theory

Stretching exponent β = 0.48, independent of T