Nuclear Resonant Scattering with Synchrotron Radiation

Svetoslav Stankov

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Outlook:

I. The Mössbauer effect.

II. Nuclear forward scattering. Comparison with the classical Mössbauer spectroscopy.

III. Nuclear inelastic scattering.

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The Mössbauer effect:

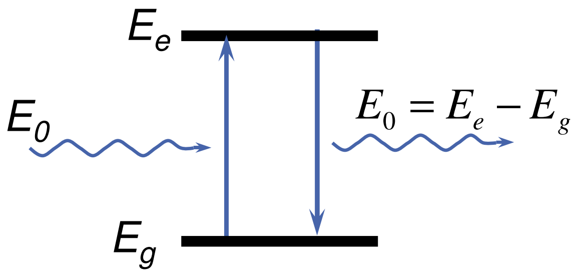
Nuclear resonant recoilless absorption/emission of γ – rays.

Nucleus of ^{57}Fe:

E_0 = 14.413 keV

τ = 141.1 ns; **Γ = 4.66 neV:**

Resolving power E_0 / Γ = ~1x10^{12}



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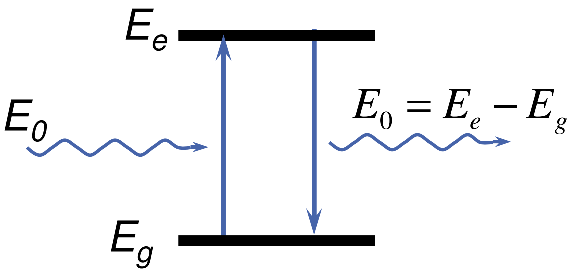
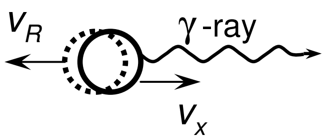


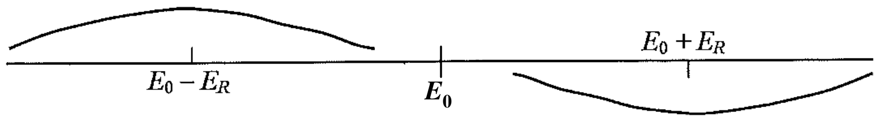


$$Mv_x = \frac{E_\gamma}{c} + M(v_x - v_R)$$

$$E_e + \frac{1}{2}Mv_x^2 = E_g + E_\gamma + \frac{1}{2}M(v_x - v_R)^2$$

Recoil energy:

$$E_R = \frac{1}{2}Mv_R^2 = \frac{E_\gamma^2}{2Mc^2} \cong 2meV$$



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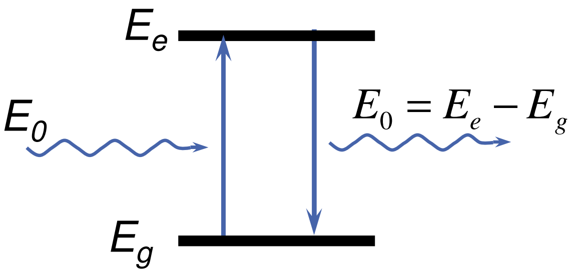

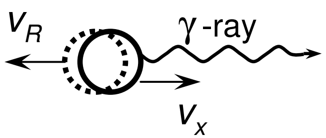

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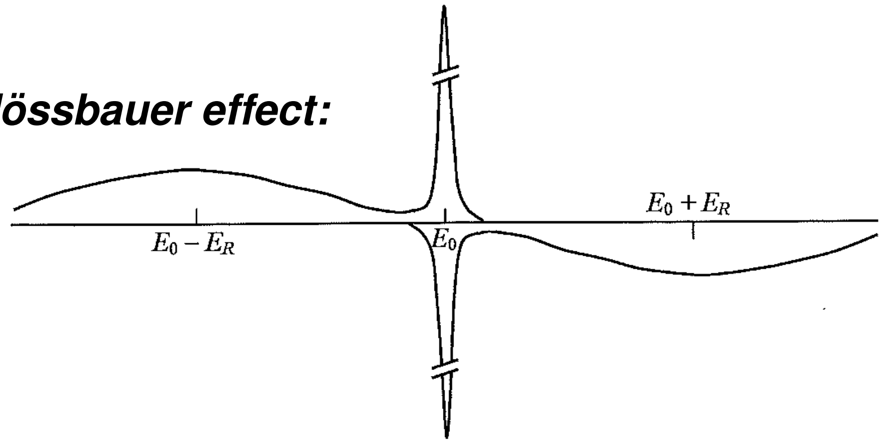

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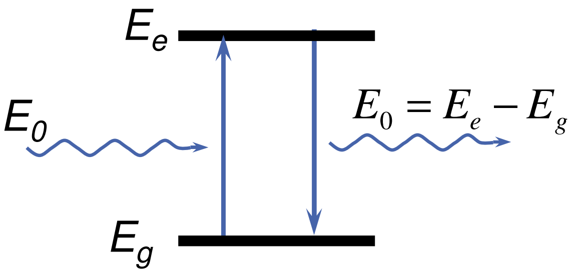
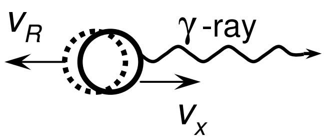
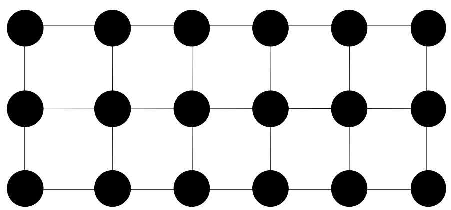
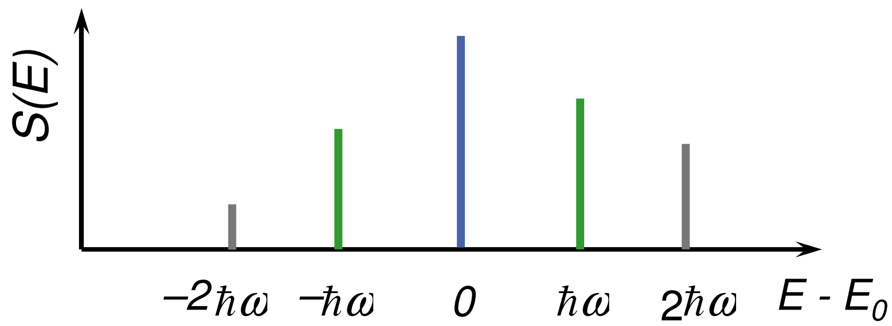






Atoms bound in a crystal lattice

Einstein model of a solid



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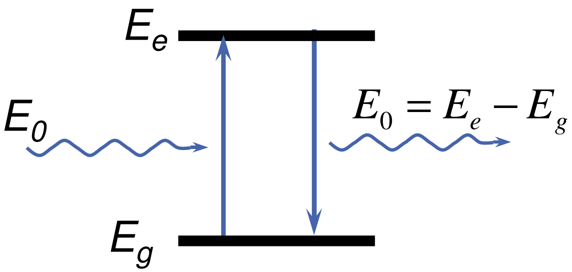
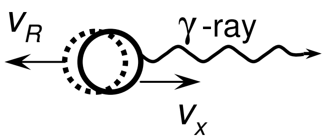
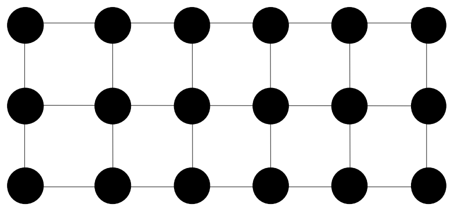


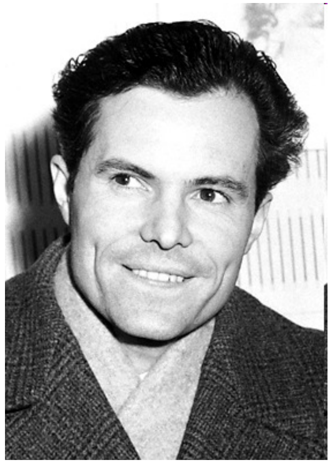




Atoms bound in a crystal lattice

S(E)

The Nobel Prize in Physics 1961
Robert Hofstadter, **Rudolf Mössbauer**



Prize motivation:
"for his researches concerning the resonance absorption of gamma radiation and his discovery in this connection of the effect which bears his name"

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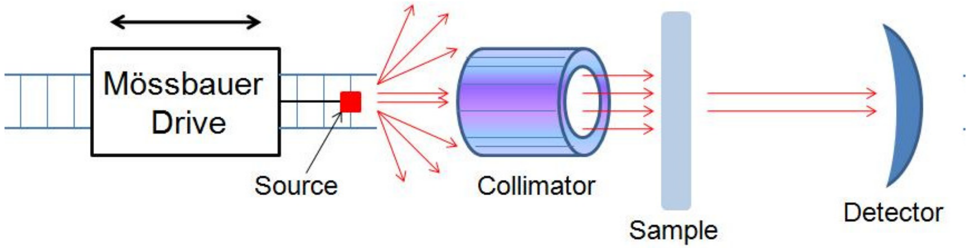
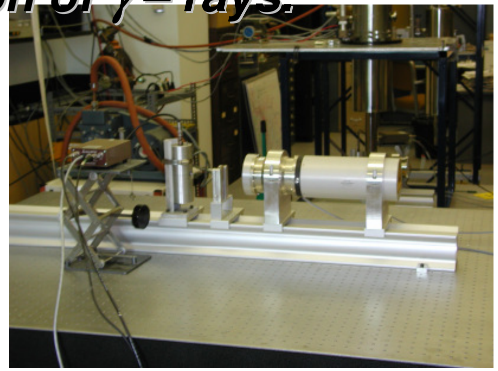
The Mössbauer effect:

Nuclear resonant recoilless absorption/emission of γ – rays.





laboratory setup

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The Mössbauer effect:

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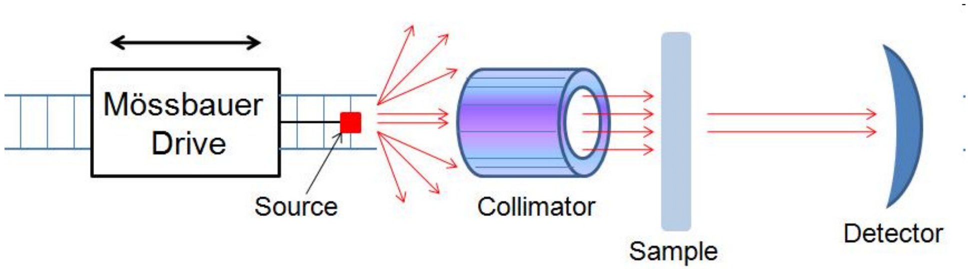

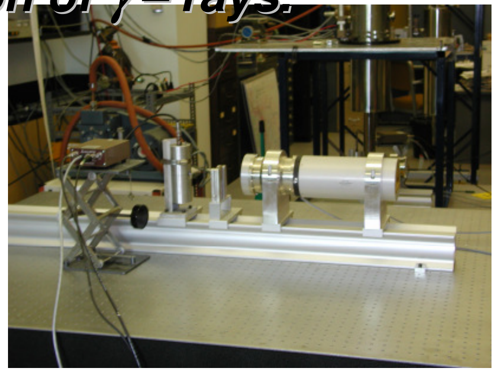

laboratory setup

PETRA III (Germany)

ESRF(France)

Spring8 (Japan)

APS (USA)

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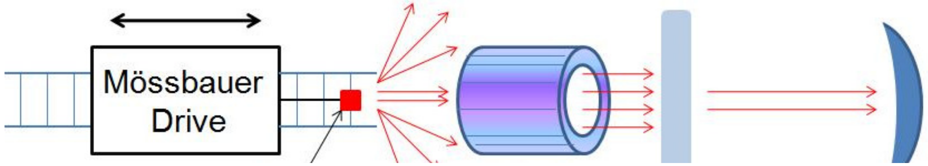
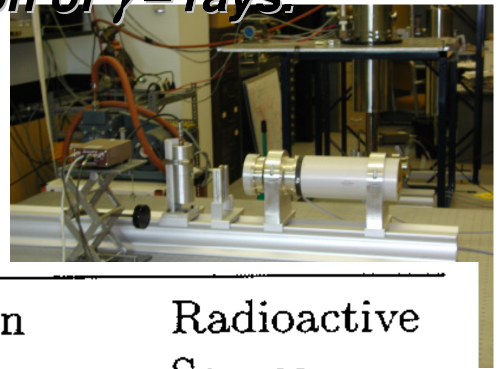
Nuclear resonant recoilless absorption/emission of γ – rays.





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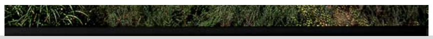
PETRA III

Property	Synchrotron Radiation	Radioactive Source
Spectral flux (ph/s/eV)	2.5×10^{12}	2.5×10^{10}
Brightness (ph/s/(eV· sr)	2.8×10^{22}	2.5×10^{13}
Brilliance (ph/s/(eV· sr ·mm^2)	2.8×10^{22}	2.5×10^{11}
Typical beam size (mm^2)	1×1	10×10
Focused beam size (µm^2)	6×6	—
Energy resolution (neV)	—	4.7
Time resolution (ns)	0.7	—
Polarization	linear or circular	unpolarized

Spr...

JSA)



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Hyperfine interactions in the nucleus of ^{57}Fe (E_e = 14.4 keV, τ = 141ns)

I. Isomer (chemical) shift:

$$\delta = E_A - E_S = \frac{2\pi}{3} zS'(z)e\Delta\rho(0)\Delta\langle r^2\rangle$$

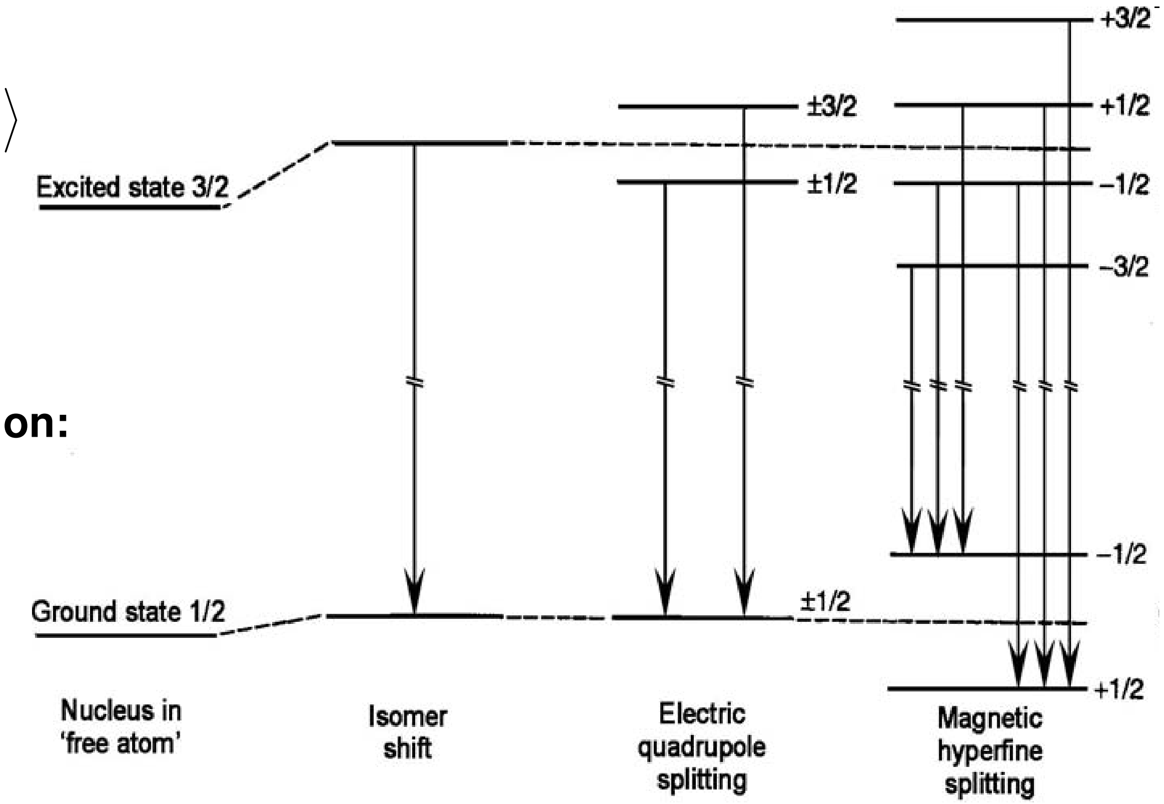
$$\Delta\langle r^2\rangle = \langle r^2\rangle_e - \langle r^2\rangle_g$$

$$\Delta\rho(0) = e\left(|\Psi_a(0)|^2 - |\Psi_s(0)|^2\right)$$

II. Electric quadrupole interaction:

$$\Delta E_Q = \frac{eQV_{zz}}{2}\left(1 + \frac{\eta^2}{3}\right)$$

$$\eta = (V_{xx} - V_{yy})/V_{zz}$$



III. Magnetic dipole interaction:

$$H_M = -\mu \cdot B = -g\mu_N I \cdot B$$

$$E_M = -gmB\mu_N$$

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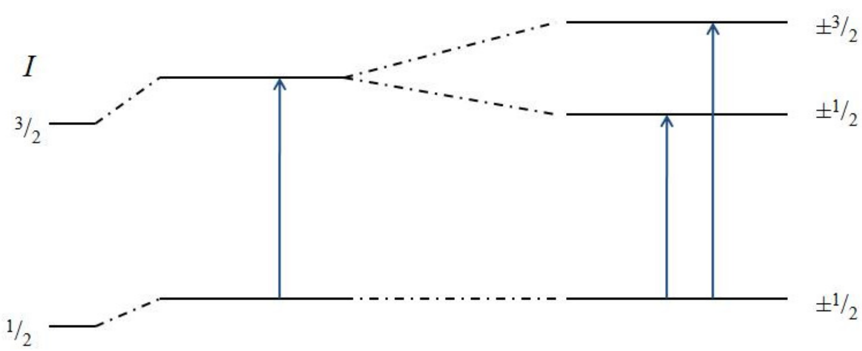
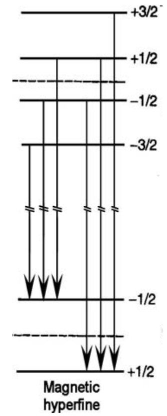
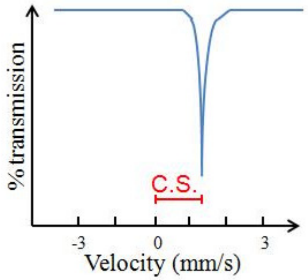
Comparison between Mössbauer and nuclear forward scattering spectra

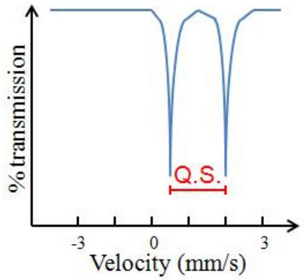
Hyperfine interactions
in the nucleus of ^{57}Fe

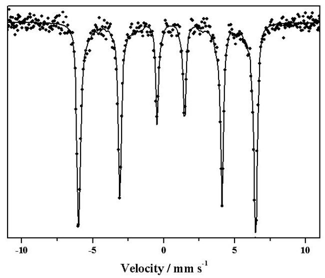




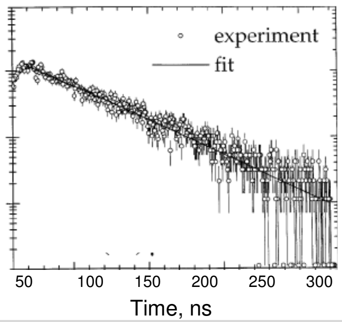
Mössbauer spectrum in the
energy domain
(classical Mössbauer spectroscopy)

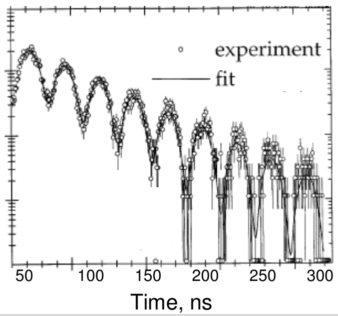


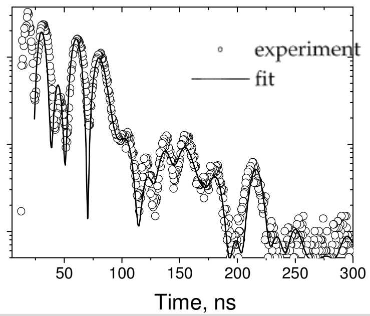




Mössbauer spectrum in the
time domain
(nuclear forward scattering)







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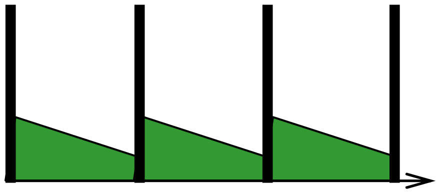
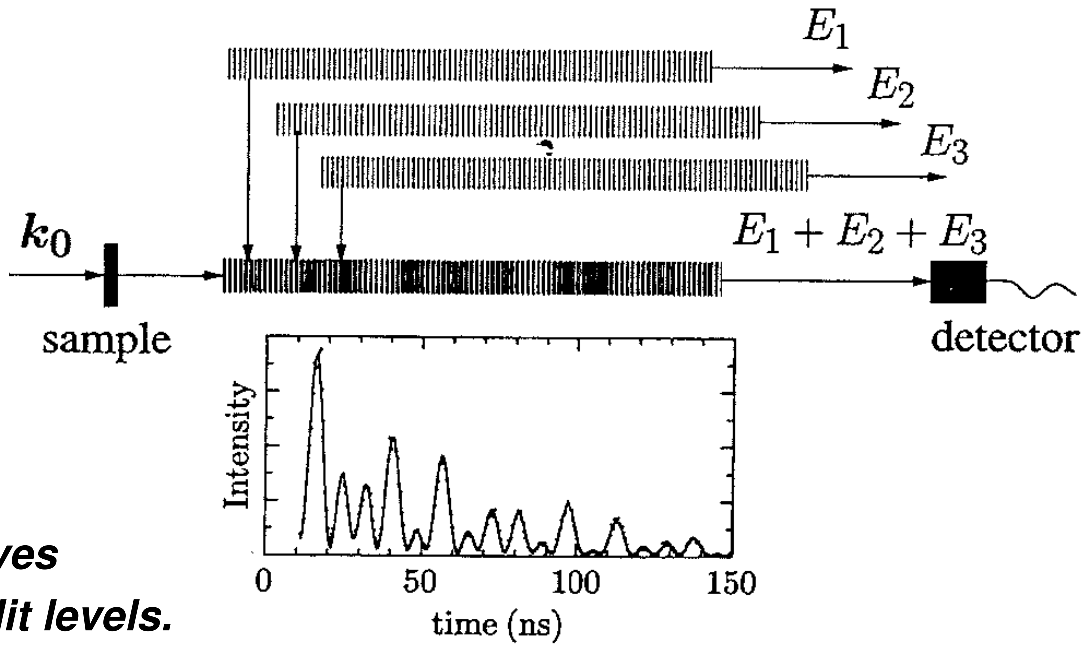
Nuclear Exciton

Simultaneous, phased in time, collective excitation of all hyperfine levels of the excited state of resonant nuclei in the sample. It propagates through the sample predominantly in spatially coherent channels (forward or Bragg direction).





Quantum beats

The coherent superposition of waves emitted from various hyperfine split levels.

R. Röhlsberger „Nuclear Condensed Matter Physics with Synchrotron Radiation" Springer 2004

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Dynamic beats

$t_a = \sigma_0 f_{LM} n_A d$ effective thickness

σ_0 maximal cross-section for nuclear resonant absorption

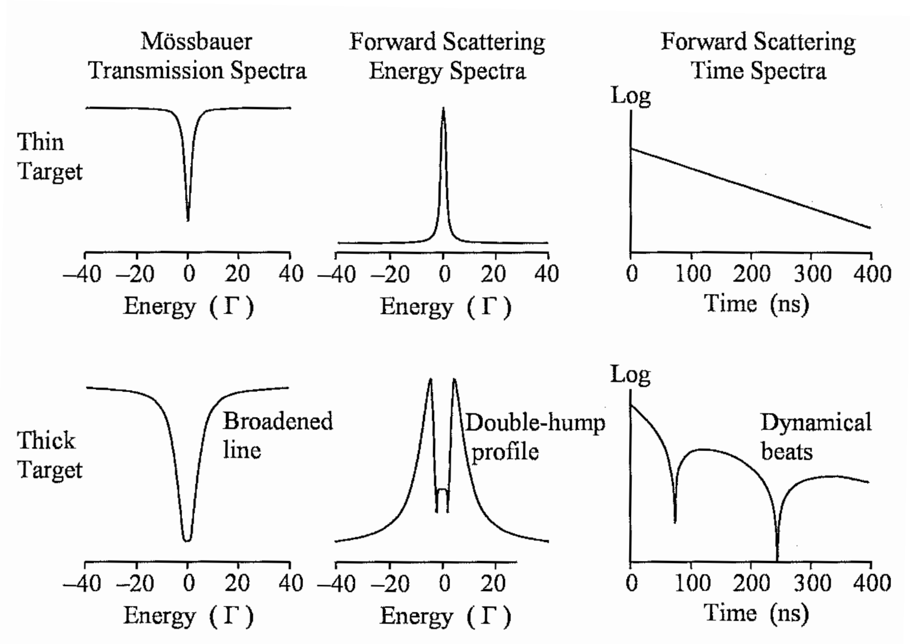
n_A Avogadro's number

f_{LM} Lamb-Mössbauer factor →

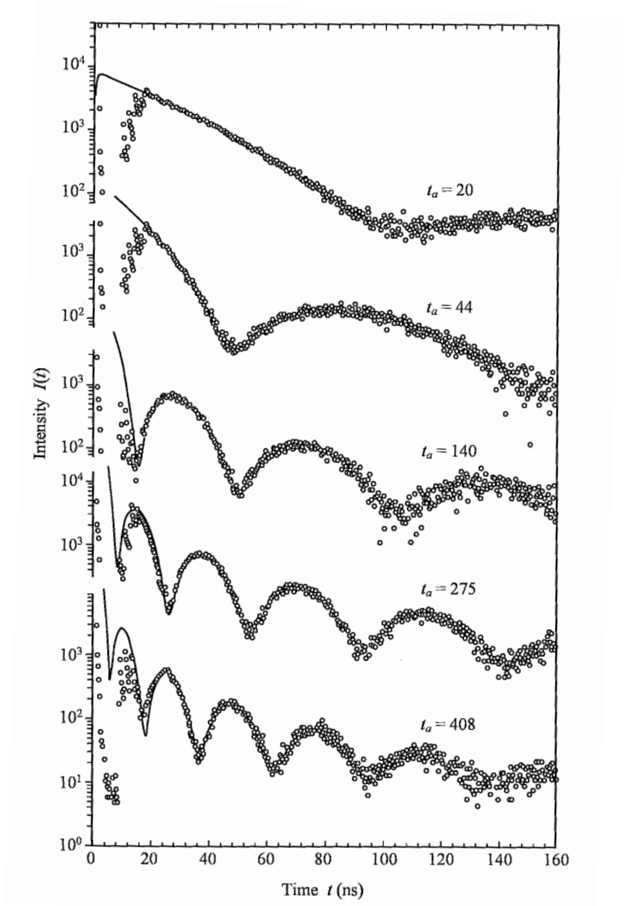
d Sample thickness

Lattice dynamics:

$$f_{LM} = e^{-k^2 \langle x^2 \rangle}$$



R. Röhlsberger „Nuclear Condensed Matter Physics with Synchrotron Radiation“ Springer 2004



Nuclear forward scattering spectra of $(NH4)_2Mg^{57}Fe(CN)_6$ for various effective thicknesses t_a

U. Van Bürck, Hyperfine Interactions ***123/124*** *483 (1999)*

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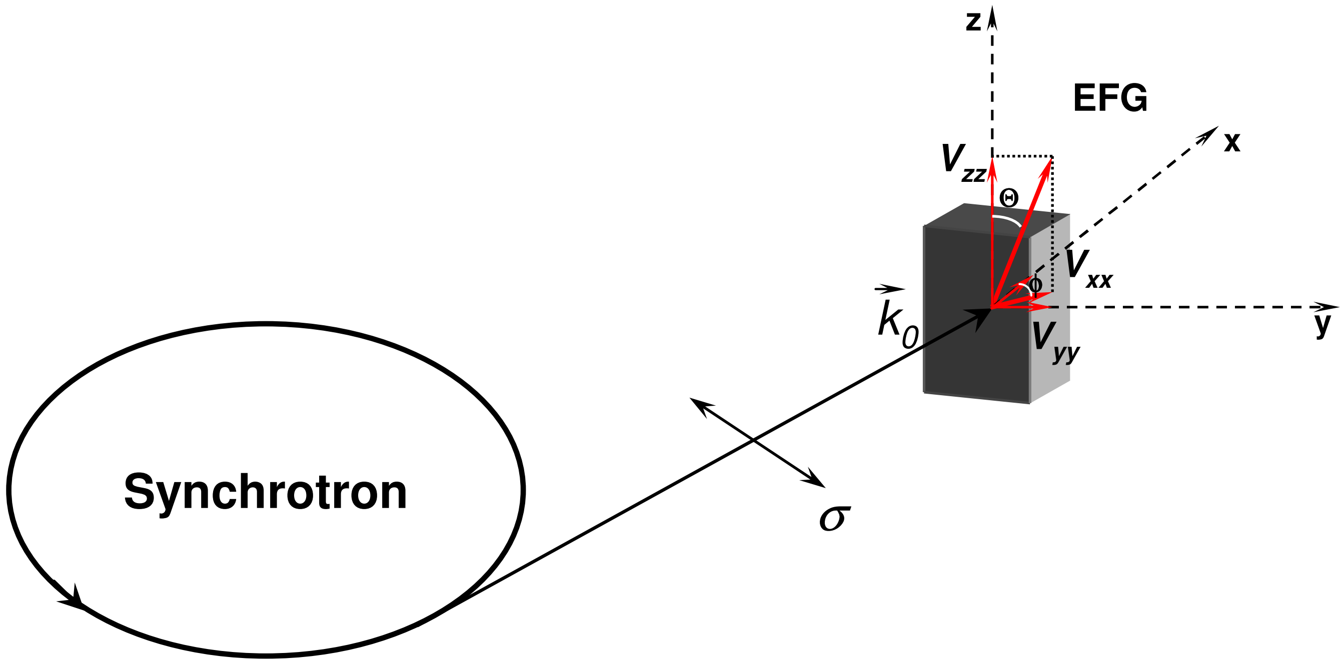
Polarization dependence of the nuclear resonant scattering



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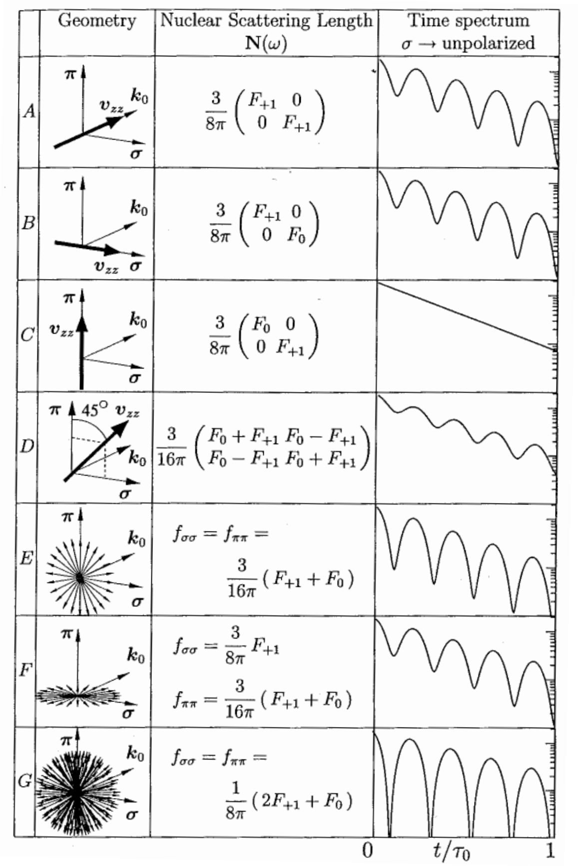
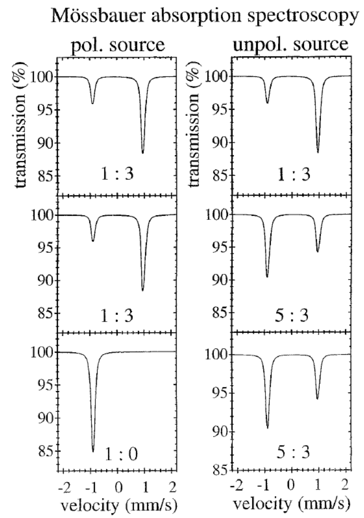
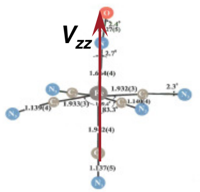
Polarization dependence of the nuclear resonant scattering

Electric quadrupole interactions

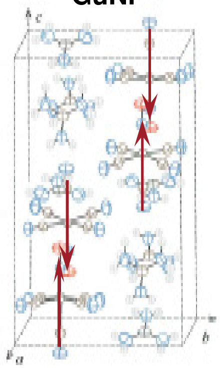


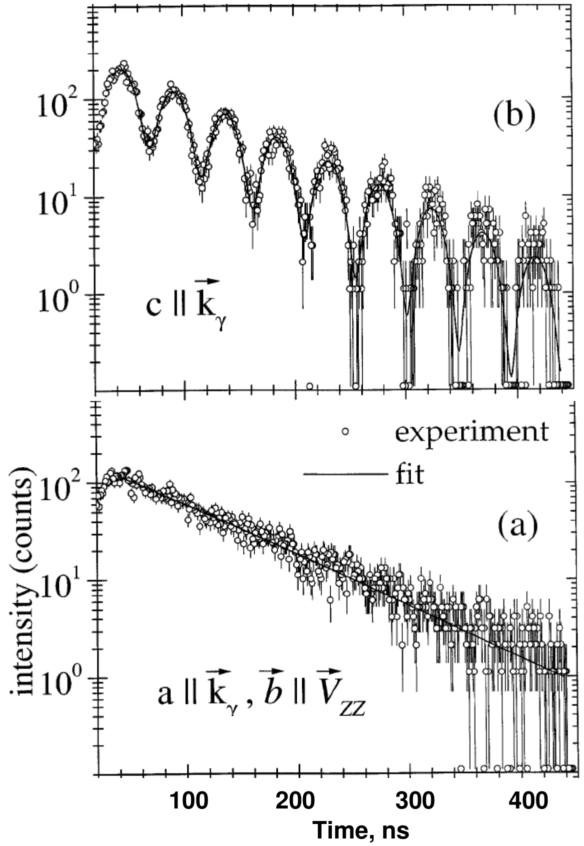


Nitropruside anion: $Fe(CN)_5NO$



GuNP





NFS spectra of $(CN3H6)_2[^{57}Fe(CN)_5NO]$ recorder at the indicaated single-crystal orientations and thicknesses.

R. Röhlsberger „Nuclear Condensed Matter Physics with Synchrotron Radiation“ Springer 2004

H. Grünsteudel et al., Hyperfine Interact. ***122****, 345 (1999)*

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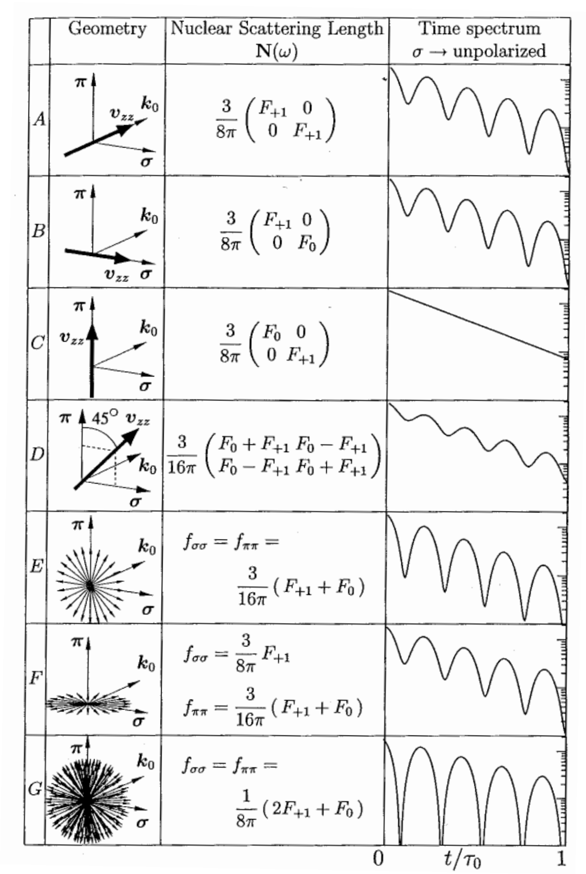
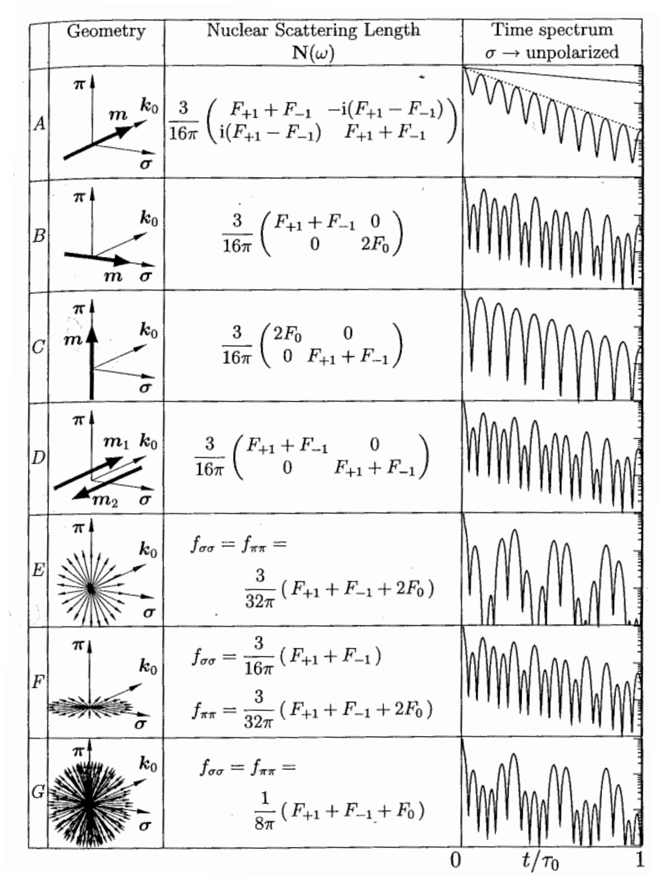
Polarization dependence of the nuclear resonant scattering

a) Electric quadrupole interactions



	Geometry	Nuclear Scattering Length N(ω)	Time spectrum σ → unpolarized
A	π, v_{zz}, k_0, σ	$\frac{3}{8\pi}\begin{pmatrix} F_{+1} & 0 \\ 0 & F_{+1} \end{pmatrix}$	
B	π, k_0, v_{zz}, σ	$\frac{3}{8\pi}\begin{pmatrix} F_{+1} & 0 \\ 0 & F_0 \end{pmatrix}$	
C	π, v_{zz}, k_0, σ	$\frac{3}{8\pi}\begin{pmatrix} F_0 & 0 \\ 0 & F_{+1} \end{pmatrix}$	
D	π, 45°, v_{zz}, k_0, σ	$\frac{3}{16\pi}\begin{pmatrix} F_0+F_{+1} & F_0-F_{+1} \\ F_0-F_{+1} & F_0+F_{+1} \end{pmatrix}$	
E	π, k_0, σ	$f_{\sigma\sigma} = f_{\pi\pi} = \frac{3}{16\pi}(F_{+1}+F_0)$	
F	π, k_0, σ	$f_{\sigma\sigma} = \frac{3}{8\pi}F_{+1}$ $f_{\pi\pi} = \frac{3}{16\pi}(F_{+1}+F_0)$	
G	π, k_0, σ	$f_{\sigma\sigma} = f_{\pi\pi} = \frac{1}{8\pi}(2F_{+1}+F_0)$	
			0 t/τ_0 1

b) Magnetic dipole interactions



	Geometry	Nuclear Scattering Length N(ω)	Time spectrum σ → unpolarized
A	π, m, k_0, σ	$\frac{3}{16\pi}\begin{pmatrix} F_{+1}+F_{-1} & -i(F_{+1}-F_{-1}) \\ i(F_{+1}-F_{-1}) & F_{+1}+F_{-1} \end{pmatrix}$	
B	π, k_0, m, σ	$\frac{3}{16\pi}\begin{pmatrix} F_{+1}+F_{-1} & 0 \\ 0 & 2F_0 \end{pmatrix}$	
C	π, m, k_0, σ	$\frac{3}{16\pi}\begin{pmatrix} 2F_0 & 0 \\ 0 & F_{+1}+F_{-1} \end{pmatrix}$	
D	π, m_1, k_0, m_2, σ	$\frac{3}{16\pi}\begin{pmatrix} F_{+1}+F_{-1} & 0 \\ 0 & F_{+1}+F_{-1} \end{pmatrix}$	
E	π, k_0, σ	$f_{\sigma\sigma} = f_{\pi\pi} = \frac{3}{32\pi}(F_{+1}+F_{-1}+2F_0)$	
F	π, k_0, σ	$f_{\sigma\sigma} = \frac{3}{16\pi}(F_{+1}+F_{-1})$ $f_{\pi\pi} = \frac{3}{32\pi}(F_{+1}+F_{-1}+2F_0)$	
G	π, k_0, σ	$f_{\sigma\sigma} = f_{\pi\pi} = \frac{1}{8\pi}(F_{+1}+F_{-1}+F_0)$	
			0 t/τ_0 1

R. Röhlsberger „Nuclear Condensed Matter Physics with Synchrotron Radiation“ Springer 2004

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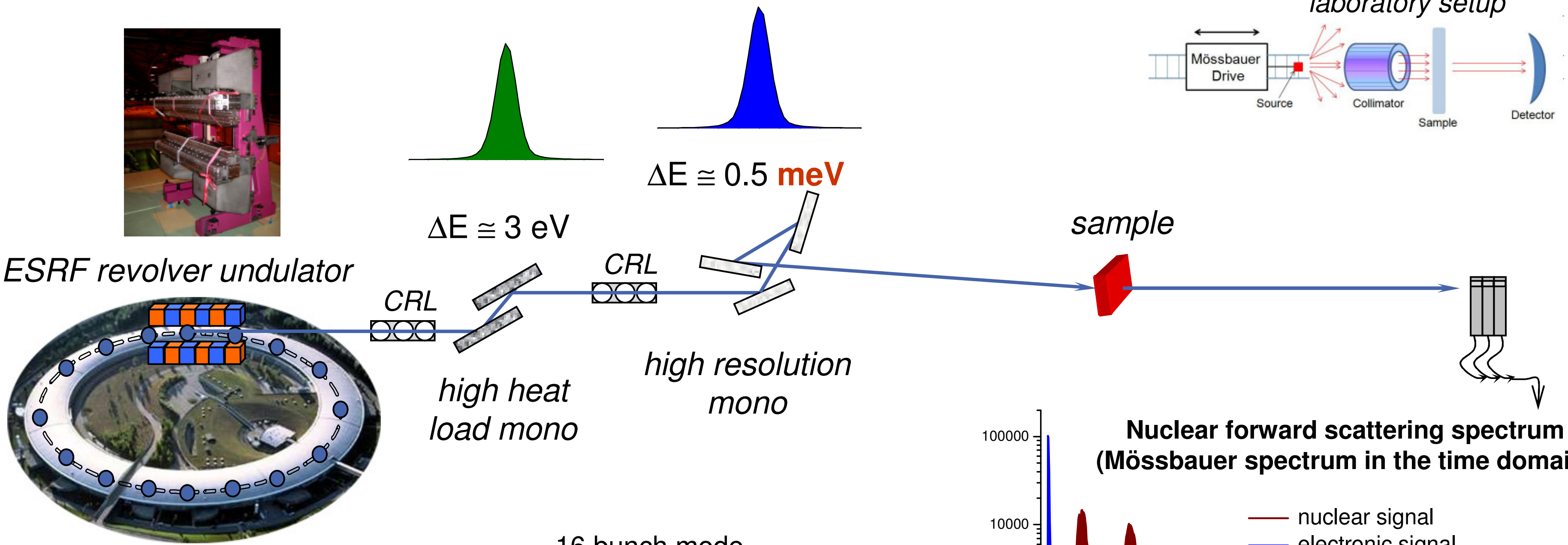
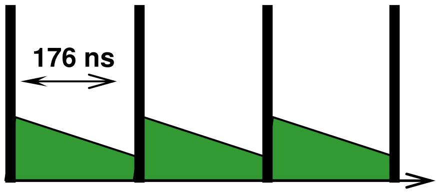
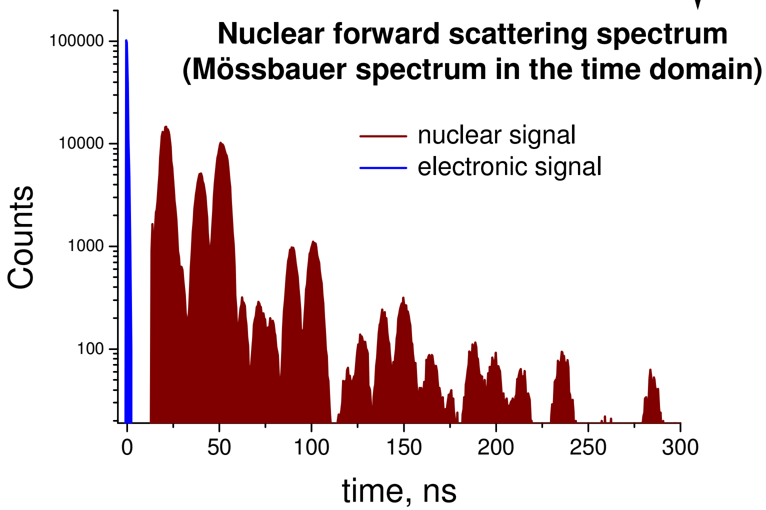
Instrumentation for nuclear resonant scattering experiments







R. Rüffer and A.I. Chumakov, Hyperfine Interact. ***97–98***, 589 (1996)

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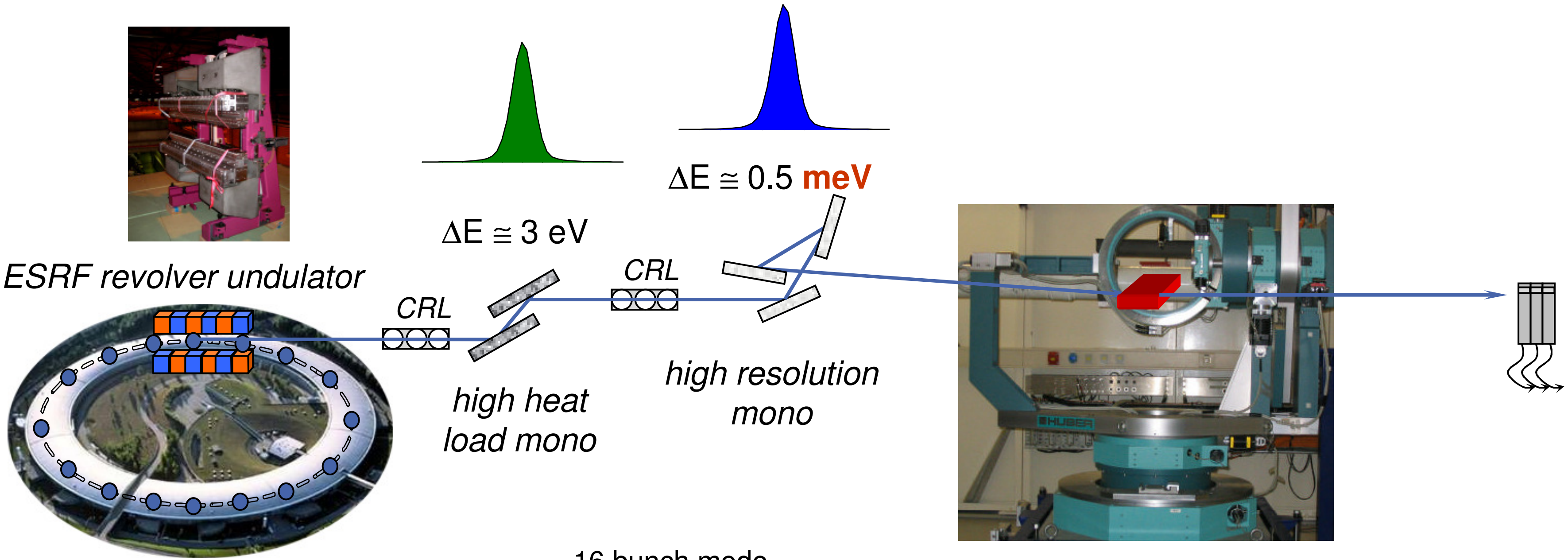
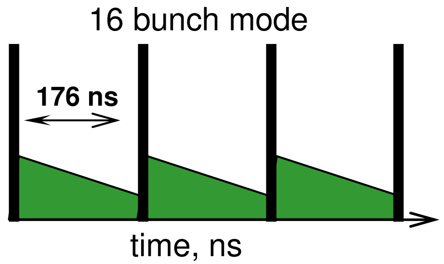
Instrumentation for nuclear resonant scattering experiments



timing mode

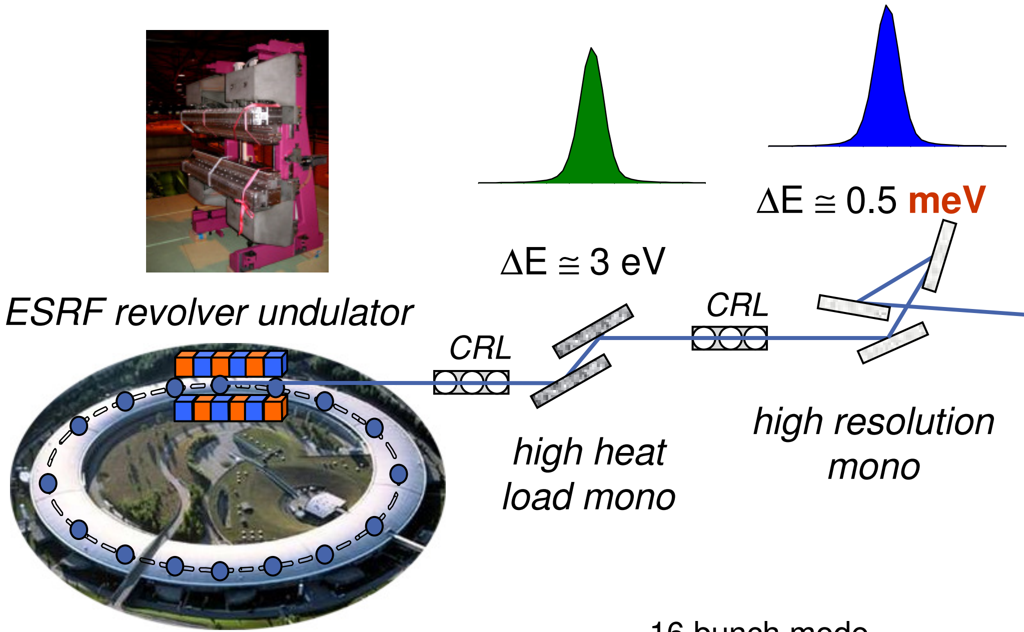


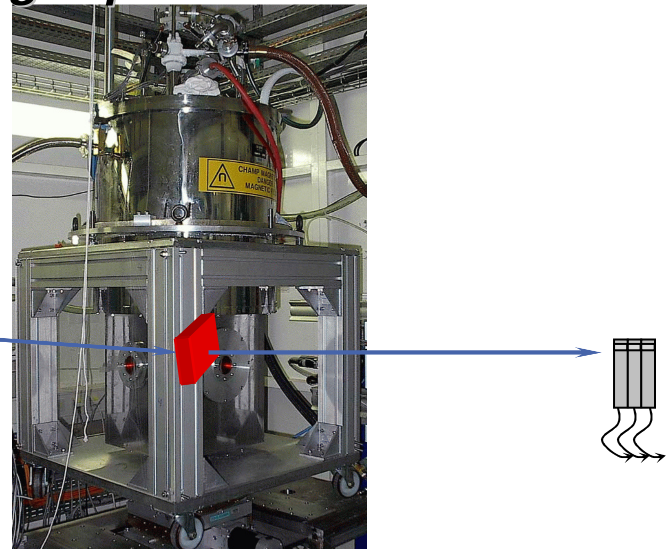
6 – cirkle diffractometer
Can accomodate:
Continuous flow cryo:300 K – 5 K
Furnace: 300 K – 1200 K

R. Rüffer and A.I. Chumakov, Hyperfine Interact. ***97–98****, 589 (1996)*

Svetoslav.Stankov@kit.edu

Instrumentation for nuclear resonant scattering experiments

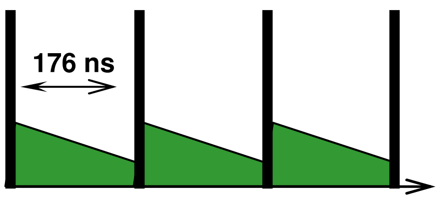




Cryo-magnet system:

Temp. range: 300 K-2 K

Magnetic field: 0 T -8 T



R. Rüffer and A.I. Chumakov, Hyperfine Interact. ***97–98****, 589 (1996)*

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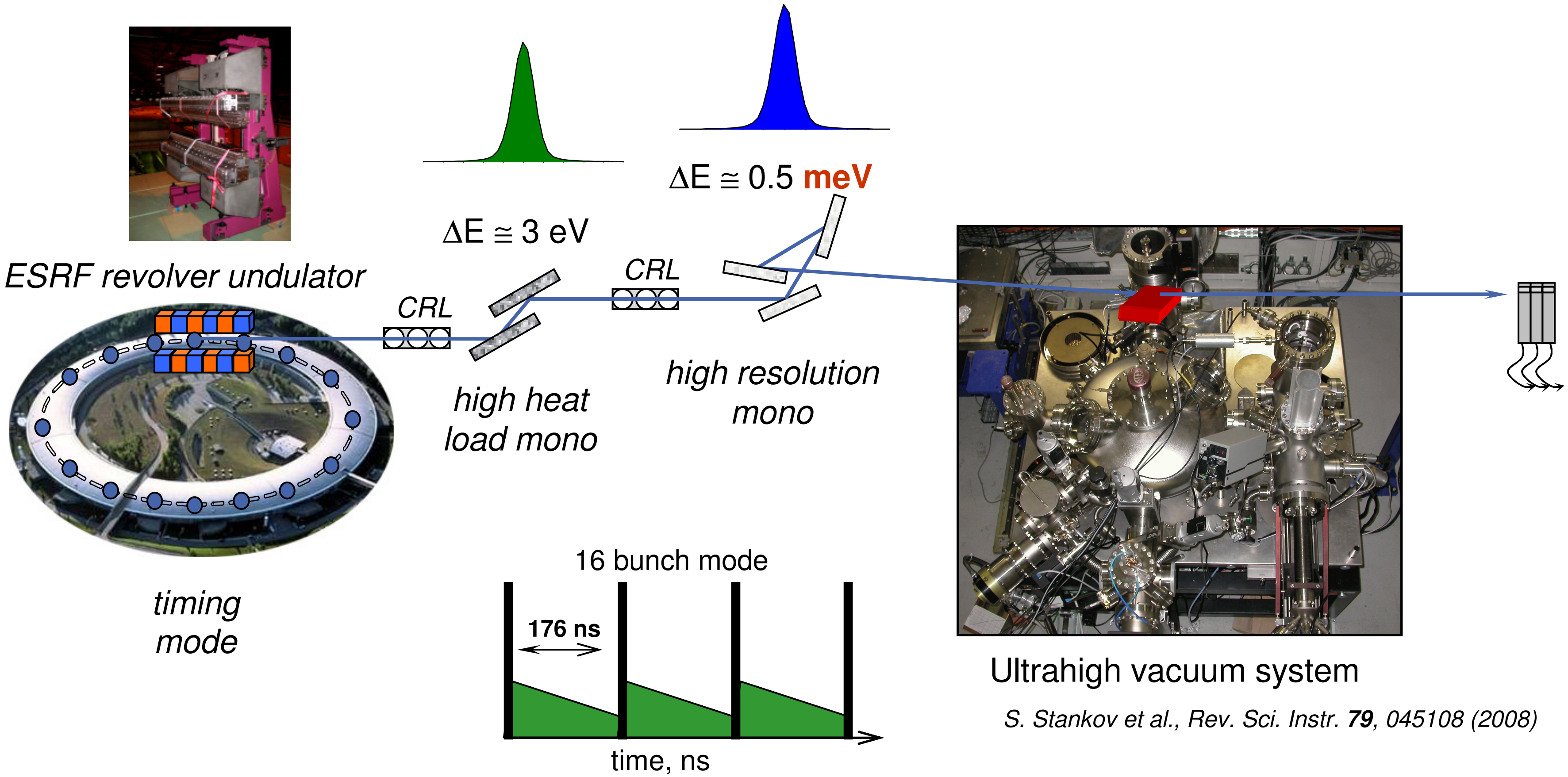
Instrumentation for nuclear resonant scattering experiments



S. Stankov et al., Rev. Sci. Instr. ***79****, 045108 (2008)*

R. Rüffer and A.I. Chumakov, Hyperfine Interact. ***97–98****, 589 (1996)*

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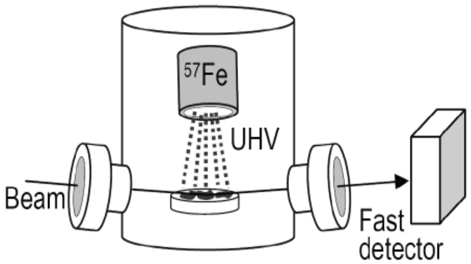
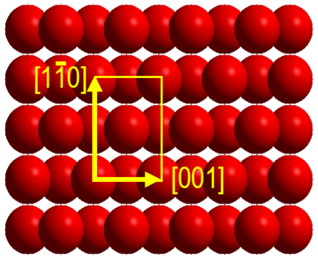
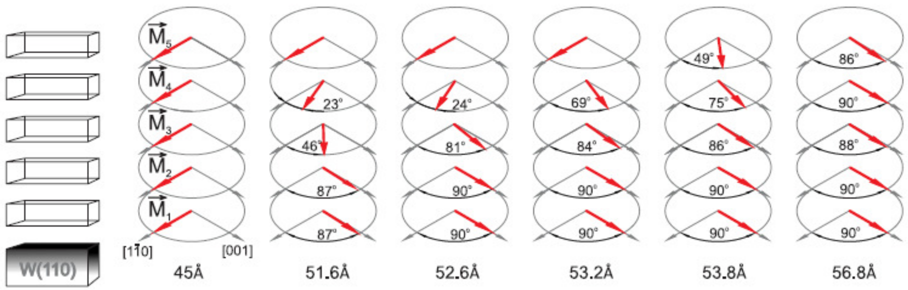
Noncollinear Magnetization Structure at the Thickness-Driven Spin-Reorientation Transition in Epitaxial Fe Films on W(110)



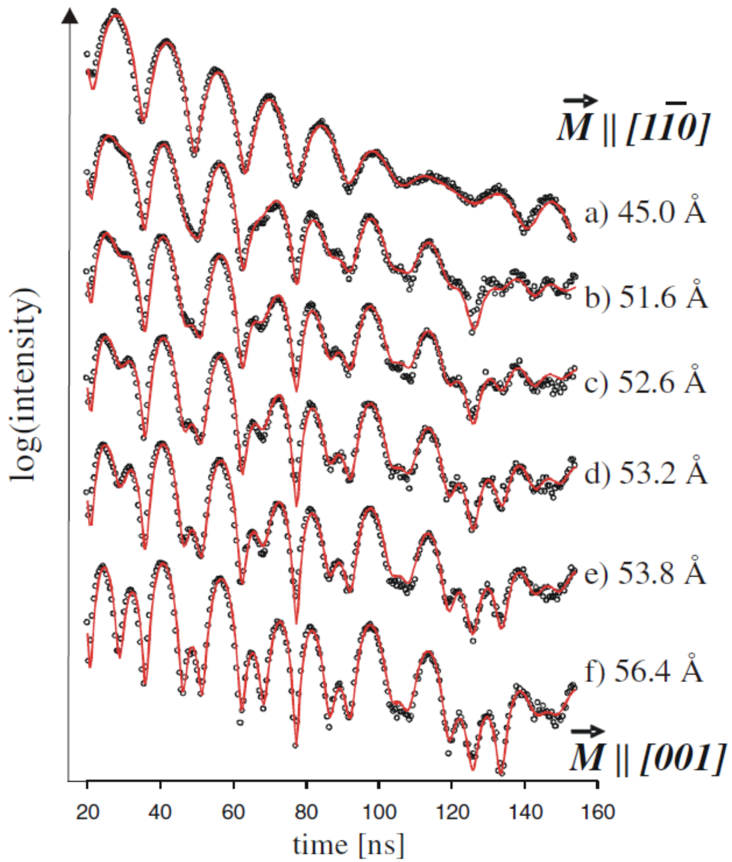


*T. Slezak et al.,J. Phys. Conf. Series **217**, 012090 (2010)*



The magnetization structure during the thickness-induced SRT for the Fe/W(110) system

*T. Slezak et al., Phys. Rev. Lett. **105**, 027206 (2010)*



NFS time spectra measured *in-situ* at the indicated film thicknesses.

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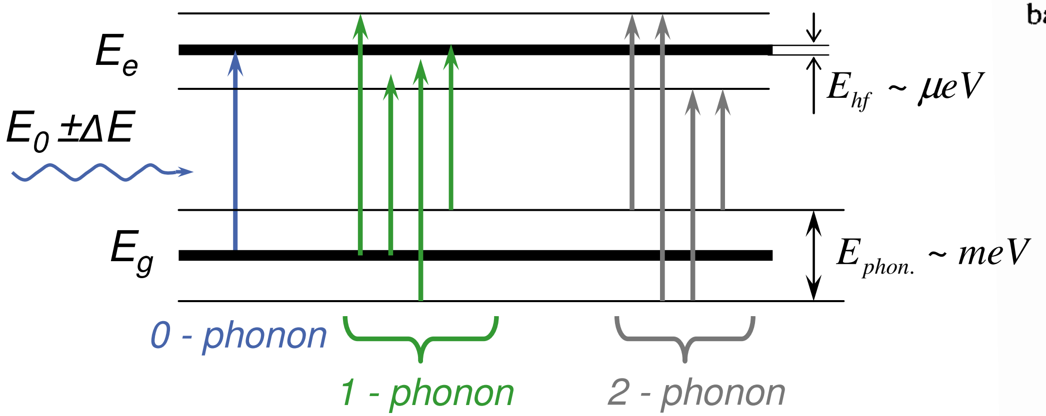
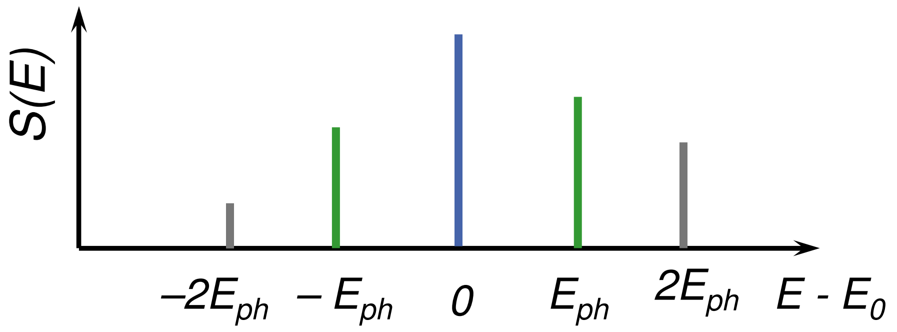
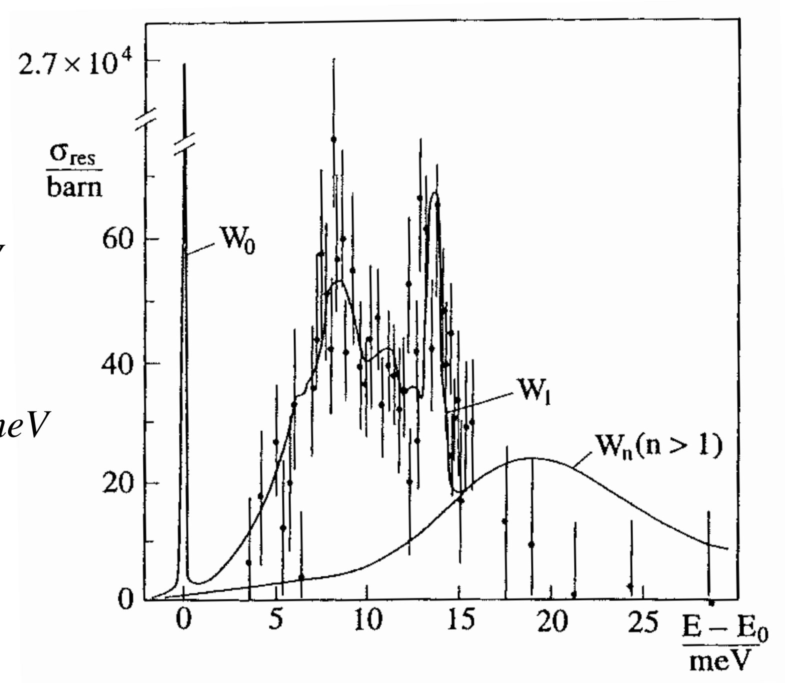
Nuclear inelastic X-ray scattering



Einstein model of a solid



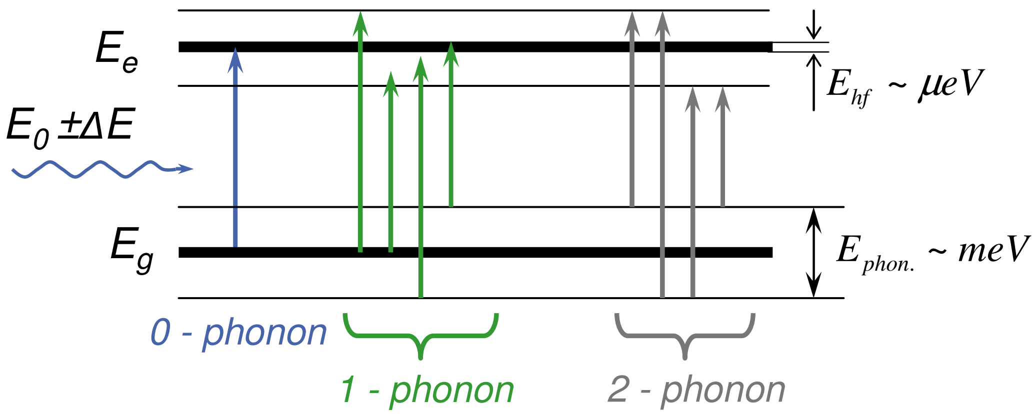


The partial phonon density of states of ^{159}Tb in TbOx (E = 58keV).

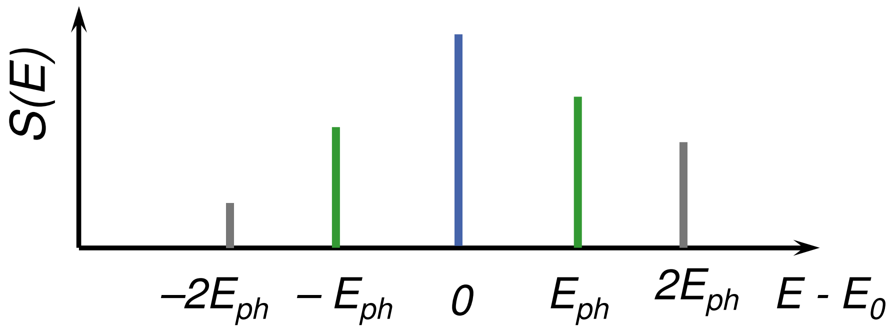
H. Weiss and H. Langhoff, Phys. Lett. ***69A****, 448 (1979)*

R. Röhlsberger „Nuclear Condensed Matter Physics with Synchrotron Radiation“

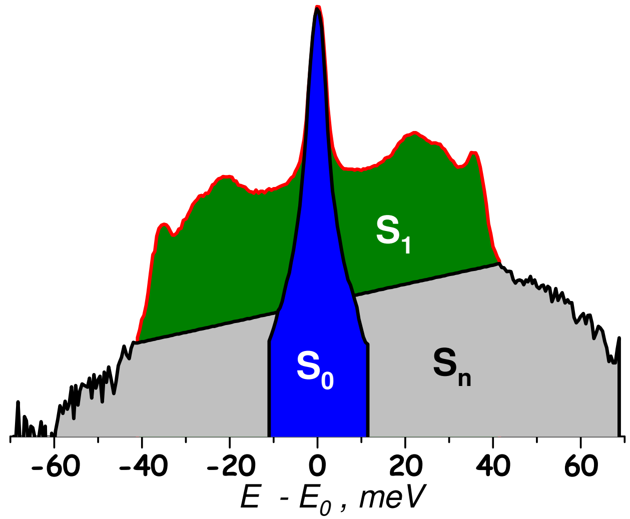
Nuclear inelastic X-ray scattering



Einstein model of a solid



R. Röhlsberger „Nuclear Condensed Matter Physics with Synchrotron Radiation“



$$W(E) = f_{LM}\left[\delta(E) + \sum_{1}^{\infty} S_n(E)\right]$$

Quasiharmonic approximation
(harmonic interaction between the atoms)

$$S_n(E) = \frac{1}{n}\int_{-\infty}^{\infty} S_1(E')S_{n-1}(E-E')dE'$$

$$S_1(E) = E_R\, g(E) / E\left(1-\exp(-E/k_BT)\right)$$

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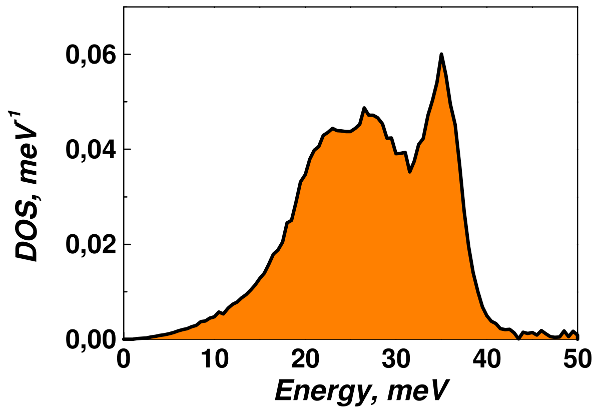
Phonon DOS determines the vibrational thermodynamics of the solid

Phonon DOS of α - Fe



- *Mean square atomic displacement* $$\langle x^2 \rangle = -\frac{\ln f_{LM}}{k_\gamma^2}$$
- *Velocity of sound* $$g(E) = \left(\frac{\tilde{m}}{m}\right)\frac{E^2}{2\pi^2\hbar^3 n v_D^3}$$
- *Vibrational contribution to the internal energy* $$U = \frac{3}{2}\int_0^\infty g(E) E \frac{e^{\beta E}+1}{e^{\beta E}-1} dE$$
- *Lattice specific heat at constant volume/pressure* $$C_V = 3k_B \int_0^\infty g(E) \frac{(\beta E)^2 e^{\beta E}}{(e^{\beta E}-1)^2} dE \qquad C_P = C_V\left(1 - T\frac{1}{v}\frac{dv}{dT}\right)$$
- *Vibrational entropy* $$S = 3k_B \int_0^\infty g(E)\left[\frac{\beta E}{2}\frac{e^{\beta E}+1}{e^{\beta E}-1} - \ln\left(e^{\beta E/2} - e^{-\beta E/2}\right)\right] dE$$
- *Mean kinetic energy and force constant* $$T(\vec{k}_\gamma) = \frac{1}{4}\int_0^\infty \tilde{g}(E,\vec{k}_\gamma) E \frac{e^{\beta E}+1}{e^{\beta E}-1} dE \qquad V(\vec{k}_\gamma) = \frac{M}{\hbar^2}\int_0^\infty \tilde{g}(E,\vec{k}_\gamma) E^2 dE$$

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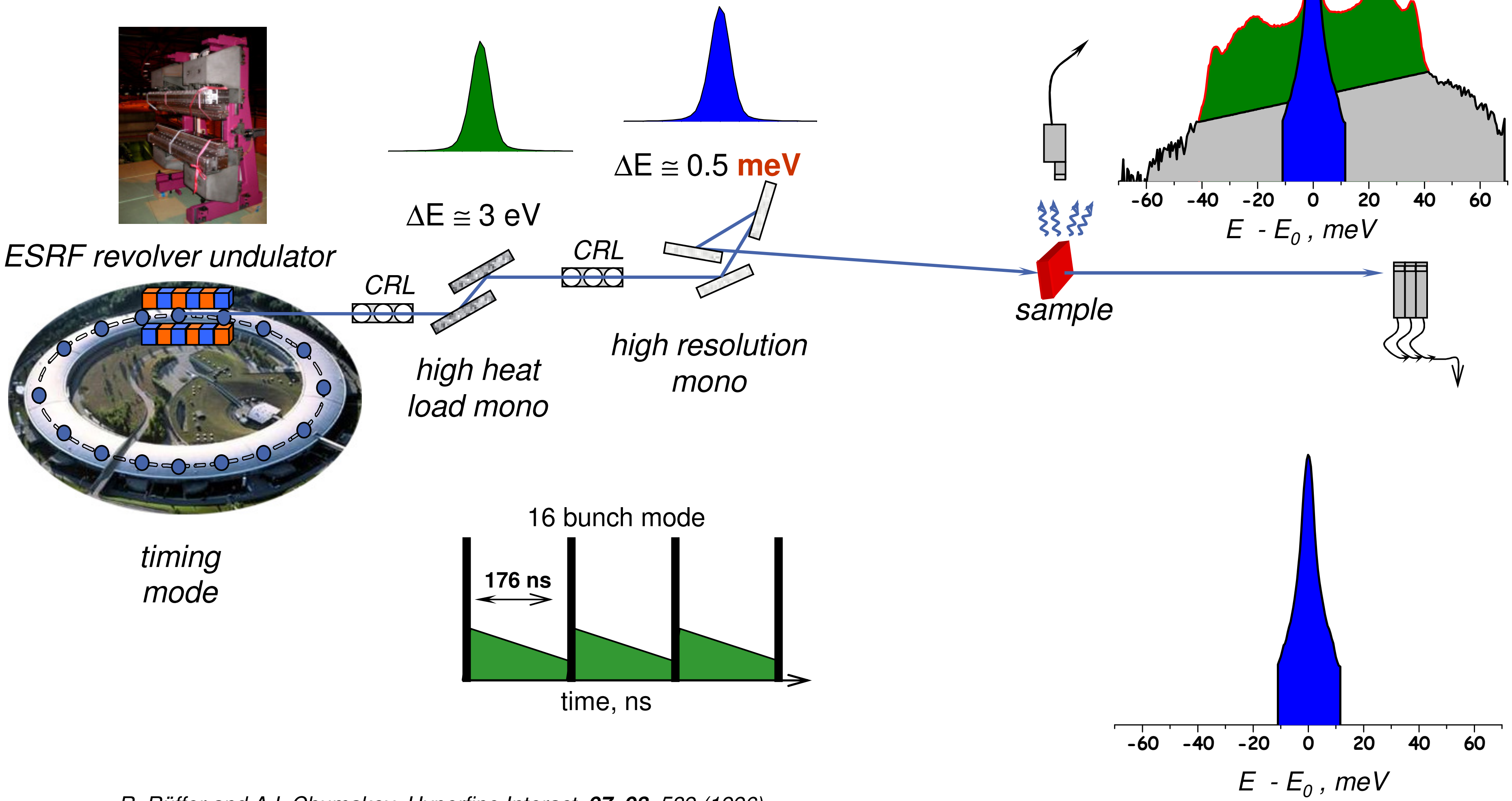
Instrumentation for nuclear inelastic scattering experiments



R. Rüffer and A.I. Chumakov, Hyperfine Interact. ***97–98****, 589 (1996)*

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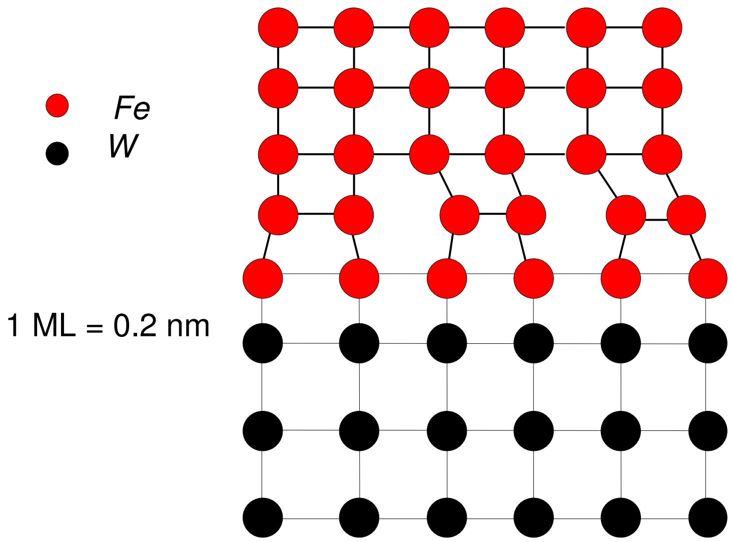
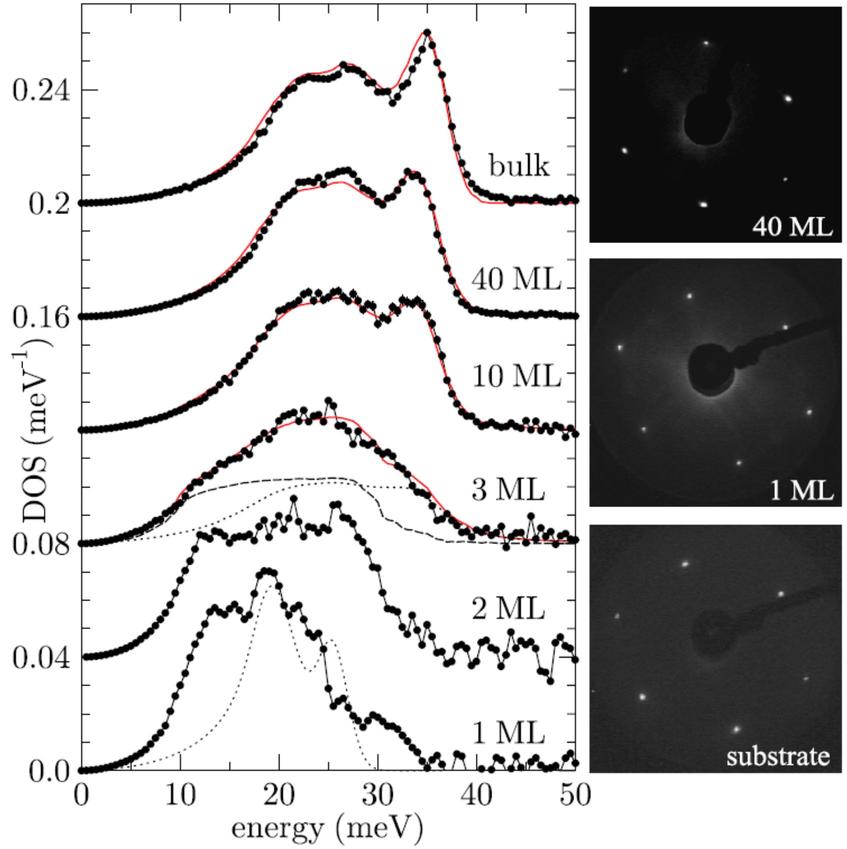
Phonons in Fe: from bulk to a monolayer

Fe/W(110)

a model system for investigation of structure, diffusion and magnetic properties of nanostructures.





misfit parameter: $\varepsilon = (a_{Fe}-a_W)/a_W$ = - 9.4 %

S. Stankov et al., Phys. Rev. Lett. ***99****, 185501 (2007)*

Summary:

- ✓ Simultaneous access to electronic, magnetic properties and lattice dynamics
- ✓ Partial (element and isotope - specific) infomration
- ✓ Access to buried layers
- ✓ Sensitive to 1 atomic layer of material
- ✓ The number of accessible isotopes is continiously increasing:

^{57}Fe, ^{119}Sn, ^{149}Sm, ^{151}Eu, ^{161}Dy, ^{83}Kr, ^{125}Te, ^{121}Sb (^{127}I,^{129}I, ^{61}Ni, ^{169}Tm ...)

- ✓ Nuclear resonance beamlines worldwide:

Grenoble - ESRF (ID18)
Hamburg - Petra III (P01)
Argonne - APS(3-ID)
Kouto - Spring-8(BL09XU)

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