Nuclear Resonant Scattering with Synchrotron Radiation

Svetoslav Stankov
Outlook:

I. The Mössbauer effect.

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

III. Nuclear inelastic scattering.
The Mössbauer effect:

Nuclear resonant recoilless absorption/emission of γ – rays.

Nucleus of $^{57}$Fe:

$E_0 = 14.413$ keV

$\tau = 141.1$ ns; $\Gamma = 4.66$ neV:

Resolving power $E_0 / \Gamma = \sim 1 \times 10^{12}$
The Mössbauer effect:

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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:

\[ E_R = \frac{1}{2} Mv_R^2 = \frac{E_\gamma^2}{2Mc^2} \approx 2 \text{meV} \]
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\end{align*}

Recoil energy: $E_R = \frac{1}{2}Mv_R^2 = \frac{E_\gamma^2}{2Mc^2} \approx 2$meV

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Recoil energy:
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Einstein model of a solid

Atoms bound in a crystal lattice

$S(E)$

$-2h\omega$, $-h\omega$, $0$, $h\omega$, $2h\omega$, $E - E_0$
The Mössbauer effect:

Nuclear resonant recoilless absorption/emission of $\gamma$ – rays.

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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"
The Mössbauer effect:

Nuclear resonant recoilless absorption/emission of $\gamma$--rays.

laboratory setup
The Mössbauer effect:

Nuclear resonant recoilless absorption/emission of γ-rays.
The Mössbauer effect:

Nuclear resonant recoilless absorption/emission of γ-rays.

<table>
<thead>
<tr>
<th>Property</th>
<th>Synchrotron Radiation</th>
<th>Radioactive Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spectral flux (ph/s/eV)</td>
<td>$2.5 \times 10^{12}$</td>
<td>$2.5 \times 10^{10}$</td>
</tr>
<tr>
<td>Brightness (ph/s/(eV·sr))</td>
<td>$2.8 \times 10^{22}$</td>
<td>$2.5 \times 10^{13}$</td>
</tr>
<tr>
<td>Brilliance (ph/s/(eV·sr·mm²))</td>
<td>$2.8 \times 10^{22}$</td>
<td>$2.5 \times 10^{11}$</td>
</tr>
<tr>
<td>Typical beam size (mm²)</td>
<td>$1 \times 1$</td>
<td>$10 \times 10$</td>
</tr>
<tr>
<td>Focused beam size (µm²)</td>
<td>$6 \times 6$</td>
<td>—</td>
</tr>
<tr>
<td>Energy resolution (neV)</td>
<td>—</td>
<td>4.7</td>
</tr>
<tr>
<td>Time resolution (ns)</td>
<td>0.7</td>
<td>—</td>
</tr>
<tr>
<td>Polarization</td>
<td>linear or circular</td>
<td>unpolarized</td>
</tr>
</tbody>
</table>


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

I. Isomer (chemical) shift:

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

$$\Delta \langle r^2 \rangle = \langle r^2 \rangle_c - \langle r^2 \rangle_s$$

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

II. Electric quadrupole interaction:

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

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

III. Magnetic dipole interaction:

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

$$E_M = -g m B \mu_N$$
Comparison between Mössbauer and nuclear forward scattering spectra

- Mössbauer spectrum in the energy domain (classical Mössbauer spectroscopy)
- Mössbauer spectrum in the time domain (nuclear forward scattering)

Hyperfine interactions in the nucleus of $^{57}$Fe
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
Dynamic beats

\[ t_a = \sigma_0 f_{LM} n_A d \]

- \( \sigma_0 \): maximal cross-section for nuclear resonant absorption
- \( n_A \): Avogadro’s number
- \( f_{LM} \): Lamb-Mössbauer factor
- \( d \): Sample thickness

Lattice dynamics:
\[ f_{LM} = e^{-k^2 \left( \frac{x}{\sigma} \right)^2} \]

Nuclear forward scattering spectra of \((\text{NH}_4)_2\text{Mg}^{57}\text{Fe(CN)}_6\) for various effective thicknesses \(t_a\)


R. Röhlserger, “Nuclear Condensed Matter Physics with Synchrotron Radiation” Springer 2004
Polarization dependence of the nuclear resonant scattering

Synchrotron
Polarization dependence of the nuclear resonant scattering

Electric quadrupole interactions

\[ \frac{3}{8\pi} \left( \frac{F_{zz} 0 \text{ } 0 F_{zz}}{F_{xx} 0 \text{ } 0 F_{xx}} \right) \]

Mössbauer absorption spectroscopy

Box 1: \( \text{pol. source} \)

Box 2: \( \text{unpol. source} \)

NFS spectra of (CN3H6)_2[\text{57Fe(CN)5NO}] recorder at the indicated 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)
Polarization dependence of the nuclear resonant scattering

a) Electric quadrupole interactions

\[ f_{ee} = f_{ee'} = \frac{3}{16\pi} (F_{e1} + F_{e2}) \]

b) Magnetic dipole interactions

\[ f_{me} = f_{me'} = \frac{3}{8\pi} (F_{m1} + F_{m2}) \]
\[ f_{me} = f_{me'} = \frac{3}{8\pi} (F_{m1} + F_{m2} + 2K_0) \]

R. Röhlsberger „Nuclear Condensed Matter Physics with Synchrotron Radiation“ Springer 2004
Instrumentation for nuclear resonant scattering experiments

ESRF revolver undulator

\[ \Delta E \cong 3 \text{ eV} \]

\[ \Delta E \cong 0.5 \text{ meV} \]

high heat load mono

high resolution mono

16 bunch mode

176 ns
time, ns

Counts

Nuclear forward scattering spectrum (Mössbauer spectrum in the time domain)

Instrumentation for nuclear resonant scattering experiments

ESRF revolver undulator

CRL

high heat load mono

16 bunch mode

time, ns

176 ns

6 – circle diffractometer
Can accommodate:
Continuous flow cryo: 300 K – 5 K
Furnace: 300 K – 1200 K

Instrumentation for nuclear resonant scattering experiments

ESRF revolver undulator

Cryo-magnet system:
Temp. range: 300 K - 2 K
Magnetic field: 0 T - 8 T

Instrumentation for nuclear resonant scattering experiments

ESRF revolver undulator

CRL CRL

high heat load mono

high resolution mono

16 bunch mode

176 ns

time, ns

Ultra high vacuum system


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


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


NFS time spectra measured in-situ at the indicated film thicknesses.
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. Röhlisberger „Nuclear Condensed Matter Physics with Synchrotron Radiation“
**Nuclear inelastic X-ray scattering**

\[ E_e \]

\[ E_0 \pm \Delta E \]

\[ E_g \]

0 - phonon

1 - phonon

2 - phonon

\[ E_{\text{phon.}} \sim \text{meV} \]

\[ E_{\text{hf}} \sim \mu\text{eV} \]

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

**Quasiharmonic approximation** (harmonic interaction between the atoms)

\[ S_{n}(E) = \frac{1}{n} \int_{-\infty}^{\infty} S_{1}(E')S_{n-1}(E - E')dE' \]

\[ S_{1}(E) = E_R g(E)/E(1 - \exp(-E/k_B T)) \]

*R. Röhlsberger „Nuclear Condensed Matter Physics with Synchrotron Radiation“*
**Phonon DOS determines the vibrational thermodynamics of the solid**

- Mean square atomic displacement
  \[ \langle x^2 \rangle = - \frac{\ln f_{LM}}{k_r^2} \]

- 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
  \[ U = \frac{3}{2} \int_0^\infty g(E) E \left( \frac{e^{\beta E}}{e^\beta} + 1 \right) \frac{e^{\beta E}}{e^\beta - 1} dE \]

- Lattice specific heat at constant volume/pressure
  \[ C_V = 3k_B \int_0^\infty g(E) \left( \frac{\beta E}{e^\beta - 1} \right)^2 \frac{e^{\beta E}}{e^\beta} dE \]
  \[ C_P = C_V \left( 1 - T \frac{1}{V} \frac{dV}{dT} \right) \]

- Vibrational entropy
  \[ S = 3k_B \int_0^\infty g(E) \left[ \frac{\beta E}{2} \frac{e^{\beta E}}{e^\beta - 1} + 1 - \ln(e^{\beta E/2} - e^{-\beta E/2}) \right] dE \]

- Mean kinetic energy and force constant
  \[ T(k_r) = \frac{1}{4} \int_0^\infty \tilde{g}(E,k_r) E \left( \frac{e^{\beta E}}{e^\beta} + 1 \right) \frac{e^{\beta E}}{e^\beta - 1} dE \]
  \[ V(k_r) = \frac{M}{\hbar^2} \int_0^\infty \tilde{g}(E,k_r) E^2 dE \]
Instrumentation for nuclear inelastic scattering experiments

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$\Delta E \approx 3$ eV

$\Delta E \approx 0.5$ meV

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176 ns

time, ns

sample

$E - E_0$, meV

$E - E_0$, meV

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: $\epsilon = (a_{Fe} - a_{W})/a_{W} = -9.4\%$

Summary:

- Simultaneous access to electronic, magnetic properties and lattice dynamics
- Partial (element and isotope - specific) information
- Access to buried layers
- Sensitive to 1 atomic layer of material
- The number of accessible isotopes is continuously increasing:
  - $^{57}\text{Fe}$, $^{119}\text{Sn}$, $^{149}\text{Sm}$, $^{151}\text{Eu}$, $^{161}\text{Dy}$, $^{83}\text{Kr}$, $^{125}\text{Te}$, $^{121}\text{Sb}$ (127I, 129I, 61Ni, 169Tm ...)
- Nuclear resonance beamlines worldwide:
  - Grenoble - ESRF (ID18)
  - Argonne - APS(3-ID)
  - Hamburg - Petra III (P01)
  - Kouto - Spring-8(BL09XU)