Synchrotron Radiation based TDPAC
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Outline

• SR TDPAC: comparison with conventional TDPAC

• SR TDPAC: comparison with MS and NRS

• Examples of application:
  • Site-specific phonon dynamics
  • Relaxation in glasses
  • Study of QI in \( \beta \)-tin

• Conclusion
Mössbauer γ rays. Here we discuss certain new time-differential-perturbed-angular-correlation- (TDPAC) type experiments which could be performed using pulsed synchrotron sources leading to interesting new results.


Advantages of SR TDPAC:
- no chemical or electronic aftereffects
- only one nuclear transition
- in general, large contrast of the beats
Formal description of SR TDPAC for M1

TDPAC

\[ I(t) \propto e^{-t/\tau} \cdot \frac{1}{2} A_{22} \cdot P_2(\cos \theta) \cdot G_{22}(t) \]

SR TDPAC

\[ I(t) \propto e^{-t/\tau} \cdot \frac{1}{2} 2A_{22} \cdot P_2(\cos \theta) \cdot G_{22}(t) \]

<table>
<thead>
<tr>
<th>Isotope</th>
<th>transition</th>
<th>$2A_{22}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{57}\text{Fe}, ^{119}\text{Sn}$</td>
<td>$1/2 \rightarrow 3/2 \rightarrow 1/2$</td>
<td>0.5</td>
</tr>
<tr>
<td>$^{61}\text{Ni}, ^{155}\text{Gd}$</td>
<td>$3/2 \rightarrow 5/2 \rightarrow 3/2$</td>
<td>0.28</td>
</tr>
<tr>
<td>$^{121}\text{Sb}, ^{151}\text{Eu}$</td>
<td>$5/2 \rightarrow 7/2 \rightarrow 5/2$</td>
<td>0.22</td>
</tr>
<tr>
<td>$^{99}\text{Ru}$</td>
<td>$5/2 \rightarrow 3/2 \rightarrow 5/2$</td>
<td>0.02</td>
</tr>
</tbody>
</table>
**SR TDPAC on $^{57}$Fe**

**Sample:**
ferrocene enriched by $^{57}$Fe
with quadrupole splitting

\[ I(t) \propto e^{-t/\tau} \cdot \frac{1}{4} \cdot 2A_{22} \cdot P_2(\cos \theta) \cdot G_{22}(t) \]

\[ G_{22}(t) = \frac{1}{5} + \frac{4}{5} \cos \Omega t \]

\[ \mu \]

\[ \mathbf{k}_{\text{in}} \]

\[ \mathbf{k}_{\text{out}} \]

\[ \mathbf{Z} \]

\[ \mathbf{1} \]

\[ \mathbf{2} \]

\[ \mathbf{3} \]

**Sample:**
α-iron enriched by $^{57}$Fe
with magnetic splitting

\[ I(t) \propto e^{-t/\tau} \cdot \frac{1}{4} \cdot 2A_{22} \cdot R(t) \]

\[ R(t) = \frac{1}{4} + \frac{3}{4} \cos 2\omega_B t \]

\[ \mu \]

\[ \mathbf{k}_{\text{in}} \]

\[ \mathbf{k}_{\text{out}} \]

\[ \mathbf{B} \]

\[ \mathbf{Z} \]

\[ \mathbf{1} \]

\[ \mathbf{2} \]

\[ \mathbf{3} \]

---

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SR TDPAC for E2 transition

Nuclear transition: $0^+ \rightarrow 2^+$,
Cascade: $0 \rightarrow 2 \rightarrow 0$

$$I(t) \propto e^{-t/\tau} \cdot \left\{1 - 2A_{22}(t) \cdot G_{22}(t) \cdot P_2(\cos \theta) - \frac{1}{4} A_{44}(t) \cdot G_{44}(t) \cdot (P_4(\cos \theta) - \frac{1}{24} P_4^{(4)}(\cos \theta) \cos 4\phi)\right\}$$

<table>
<thead>
<tr>
<th>Element</th>
<th>$\tau$</th>
<th>$E$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{154}$Sm</td>
<td>4.3</td>
<td>82.0</td>
</tr>
<tr>
<td>$^{158}$Gd</td>
<td>3.7</td>
<td>79.5</td>
</tr>
<tr>
<td>$^{160}$Gd</td>
<td>3.9</td>
<td>75.3</td>
</tr>
<tr>
<td>$^{164}$Dy</td>
<td>3.4</td>
<td>73.4</td>
</tr>
<tr>
<td>$^{166}$Er</td>
<td>2.7</td>
<td>80.6</td>
</tr>
<tr>
<td>$^{168}$Er</td>
<td>2.7</td>
<td>79.8</td>
</tr>
<tr>
<td>$^{172}$Yb</td>
<td>2.6</td>
<td>78.7</td>
</tr>
<tr>
<td>$^{174}$Yb</td>
<td>2.5</td>
<td>76.5</td>
</tr>
<tr>
<td>$^{180}$Hf</td>
<td>2.2</td>
<td>93.3</td>
</tr>
<tr>
<td>$^{182}$W</td>
<td>1.9</td>
<td>100.1</td>
</tr>
</tbody>
</table>

Experimental setup

- Monochromator
- High Resolution Monochromator
- Al foil
- Undulator
- Sample
- Detector
- Detector with Si APDs
- Energy band width: 1-30 meV
- Dynamical range: \(~10^6\) ph/s
- Time resolution: \(~0.1\)-2 ns
Differences:
• dependence on the spatial evolution of nuclei
• dependence on the ground nuclear state spin evolution
Energy level diagram:

$^{57}$Fe, quadrupole splitting

$I=3/2$

$I=1/2$

Coherent superposition of the wavelets

$E \propto e^{i\Omega t} + e^{-i\Omega t}$
Hyp. Int.: magnetic splitting

α-iron

MS, NFS
SR TDPAC

Relative transmission

SR TDPAC

Log I(t)

Velocity [mm/s]

MS

SR

NFS
Applications

• Site-specific phonon dynamics

• Relaxation in glasses

• Study of QI in $\beta$-tin
Sample:
Magnetite, Fe$_3$O$_4$

A site: tetrahedral, $B = 49.0$ T
B site: octahedral, $B = 46.0$ T
\[ I(t) \propto e^{-t/\tau} \cdot \left( \frac{1}{4} - 2A_{22} \cdot R(t) \right) \]

\[ R(t) = A \cdot \left( \frac{1}{4} + \frac{3}{4} \cos 2\omega_A t \right) + B \cdot \left( \frac{1}{4} + \frac{3}{4} \cos 2\omega_B t \right) \]
MOLECULAR MOTIONS IN A VISCOUS ORGANIC LIQUID: FERROCENE IN COLD BUTYL PHTHALATE (*)

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Carnegie Mellon University, Pittsburgh, Pennsylvania 15213, U. S. A.

Sample:
ferrocene
Fe(C₅H₅)₂

Glass-former:
Di-butyl phthalate,

Tₙ = 178 K

Task:
resolve rotational and translational degrees of motions
Relaxation process

- $k$ – jump rate, sec$^{-1}$
- $a$ – jump distance
- $\alpha$ – jump angle
- $\lambda$ – wavelength, 0.86 Å
- $R$ – radius of the molecule, 2 Å

\[ \Delta_T = \frac{2}{3} \pi^2 \cdot k \left( \frac{a}{\lambda} \right)^2 \]

\[ \Delta_R = \frac{3}{2} \cdot k \cdot \alpha^2 \]

\[ \alpha = \sqrt{\frac{2}{3} \cdot \frac{a}{R}} \]

\[ \frac{\Delta_R}{\Delta_T} = \frac{3}{2\pi^2} \cdot \left( \frac{\lambda}{R} \right)^2 \rightarrow 0.03 \]
Relaxation in glasses

Sample:

<table>
<thead>
<tr>
<th>Sample Description</th>
<th>Temperature</th>
</tr>
</thead>
<tbody>
<tr>
<td>Di-butyl phthalate, $T_g = 178$ K</td>
<td></td>
</tr>
<tr>
<td>Di isobutyyl phthalate, $T_g = 188$ K</td>
<td></td>
</tr>
<tr>
<td>5% ferrocene + DiBP+FC</td>
<td></td>
</tr>
</tbody>
</table>

SR TDPAC

DiBP+FC

T = 180 K

SR TDPAC

DiBP+FC

NFS

SR

T = 280 K

T = 205 K

T = 215 K

NFS

Intensity

Time / ns

$T_g = 178$ K

$T_g = 188$ K

5% ferrocene + DiBP+FC

SR TDPAC

DiBP+FC

NFS

SR

Intensity

Time / ns
Conclusions:
The probe reproduces the dynamics of the glass former.
At low T dynamics of the probe follows slow $\beta$ branch
At low T only molecular rotation is seen in both NFS and SR TDPAC.
Study of $\beta$-tin by SR TDPAC

K.P. Mitrofanov et al., Sov. Phys. JETP 21 (1965) 524


Measurements by SR TDPAC
Study of $\beta$-tin by SRPAC

Results of measurements

Conclusion:
• value of the life time of 23.9keV nuclear state of $^{119}$Sn was obtained with high precision
• quadrupole splitting of $\beta$-tin have been seen in the time spectrum and was measured for different temperatures
Conclusion

SR TDPAC – method which allows to extend study of hyperfine interactions by TDPAC on to Mössbauer isotopes

– method which allows to extend study of hyperfine interactions by MS into the range of zero Lamb-Mössbauer factor.

3 main directions of application:

Complementary to MS information about hyperfine splitting and dynamics

Hyperfine interactions in high energy Mössbauer isotopes

Study of hyperfine interactions and dynamics in soft condensed matter
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