Synchrotron Radiation based TDPAC

I. Sergueev

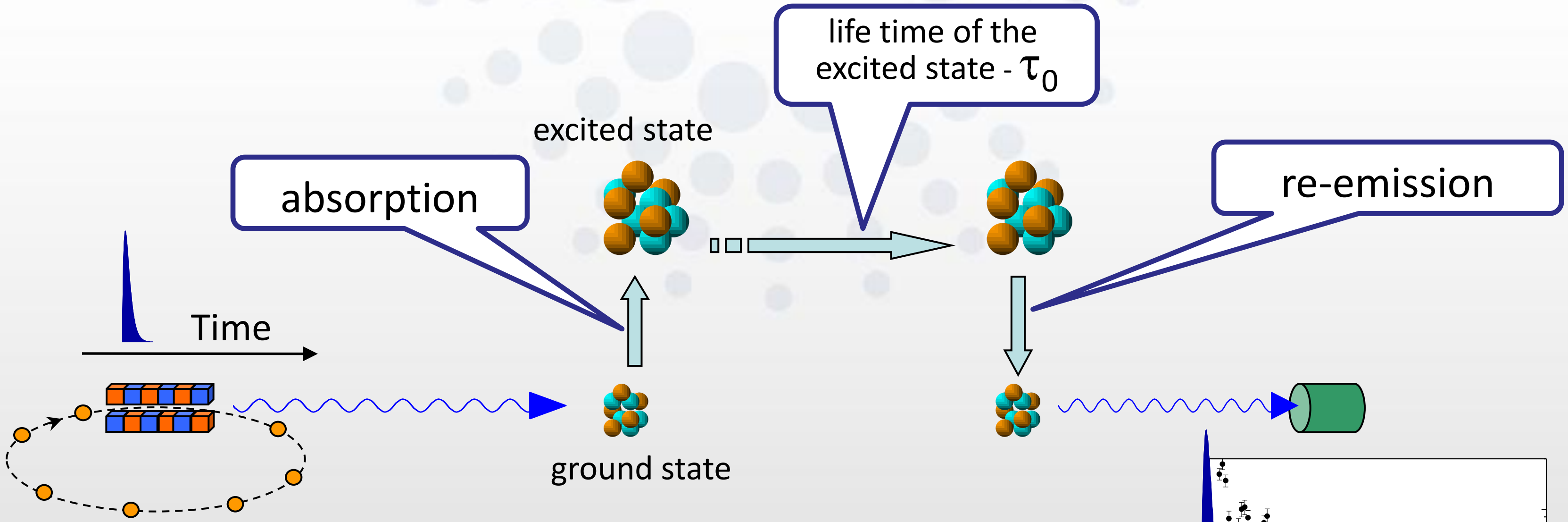
European Synchrotron Radiation Facility

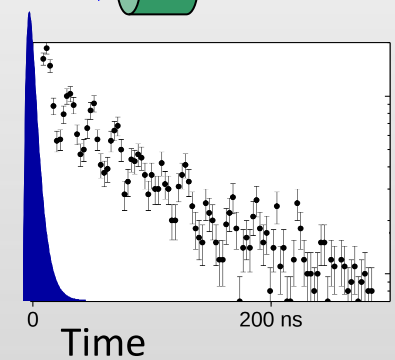
Grenoble, France

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 β-tin
- Conclusion

Nuclear Resonance Scattering





PHYSICAL REVIEW B VOLUME 18, NUMBER 1 1 JULY 1978

Quantum beats from nuclei excited by synchrotron pulses

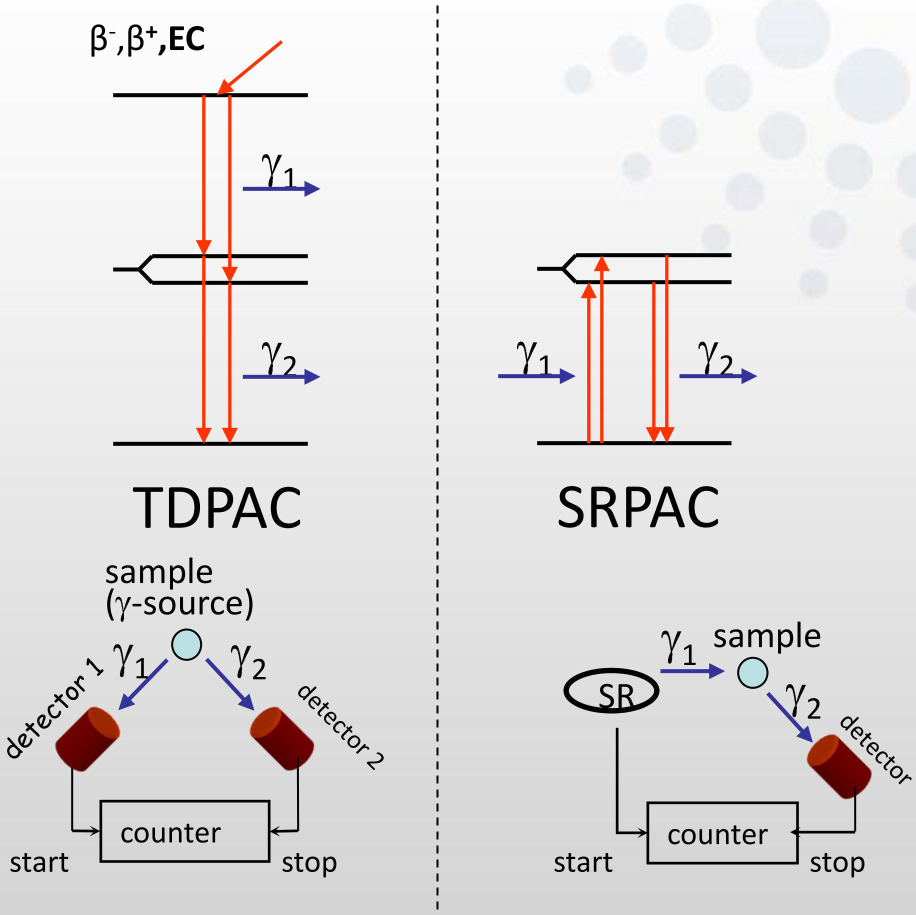
G. T. Trammell and J. P. Hannon
Physics Department, Rice University, Houston, Texas 77001
(Received 27 October 1977)

Mössbauer γ rays. Here we discuss certain new time-differential-perturbed-angular-correlation- (TDPAC) type experiments[5] which could be performed using pulsed synchrotron sources leading to interesting new results.

*A.Q.Baron et al, Europhys. Lett.,**34**, 331(1996)*

*I.Sergueev et al, Phys. Rev. B **73**, 024203 (2006)*
*I.Sergueev et al. Phys. Rev. B **78**, 214436 (2008)*

TDPAC vs SR TDPAC

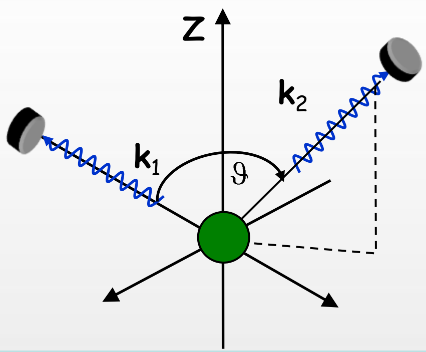


Advantages of SR TDPAC:

- no chemical or electronic aftereffects
- only one nuclear transition
- in general, large contrast of the beats

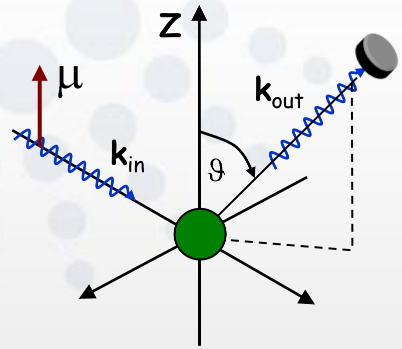
Formal description of SR TDPAC for M1

TDPAC



$$I(t) \propto e^{-t/\tau} \cdot 1 + A_{22} \cdot P_2(\cos\theta) \cdot G_{22}(t)$$

SR TDPAC



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

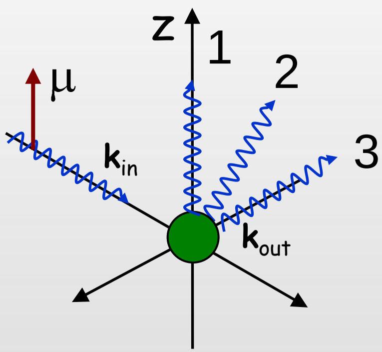
Isotope	transition	$2A_{22}$
^{57}Fe, ^{119}Sn	1/2→3/2→1/2	0.5
^{61}Ni, ^{155}Gd	3/2→5/2→3/2	0.28
^{121}Sb, ^{151}Eu	5/2→7/2→5/2	0.22
^{99}Ru	5/2→3/2→5/2	0.02

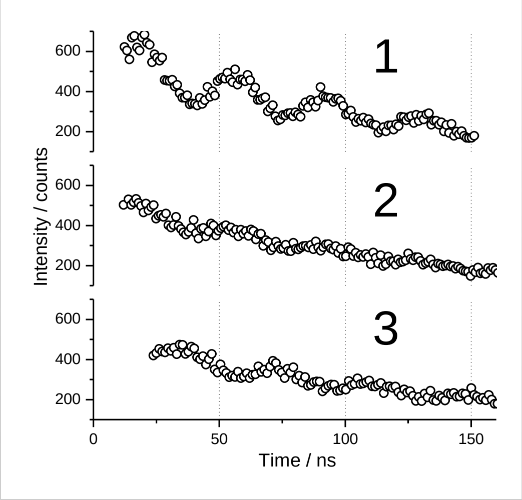
SR TDPAC on ^{57}Fe

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

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

$$G_{22}(t) = \tfrac{1}{5} + \tfrac{4}{5}\cos\Omega t$$

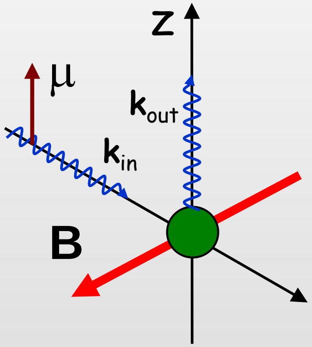


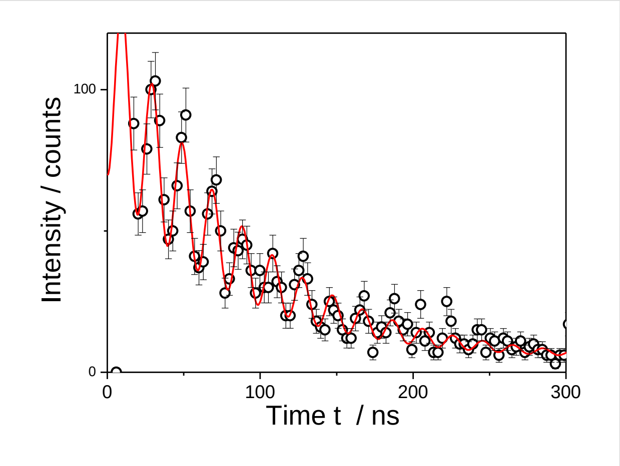


Sample:
α-iron enriched by ^{57}Fe
with magnetic splitting

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

$$R(t) = \tfrac{1}{4} + \tfrac{3}{4}\cos 2\omega_B t$$





SR TDPAC for E2 transition

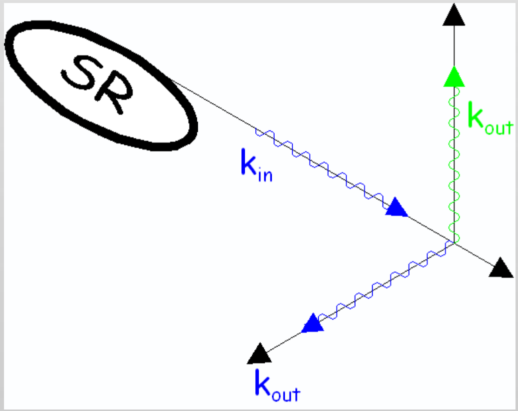
Nuclear transition : $0^+ \rightarrow 2^+$,

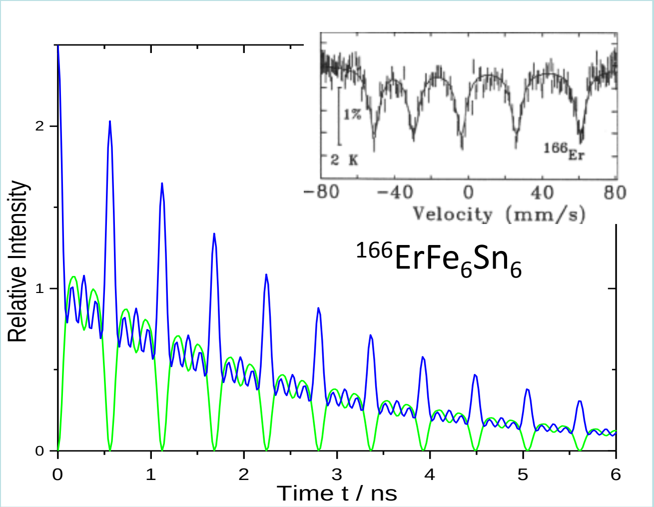
Cascade: $0 \rightarrow 2 \rightarrow 0$

	τ ns	E keV
^{154}Sm	4.3	82.0
^{158}Gd	3.7	79.5
^{160}Gd	3.9	75.3
^{164}Dy	3.4	73.4
^{166}Er	2.7	80.6
^{168}Er	2.7	79.8
^{172}Yb	2.6	78.7
^{174}Yb	2.5	76.5
^{180}Hf	2.2	93.3
^{182}W	1.9	100.1

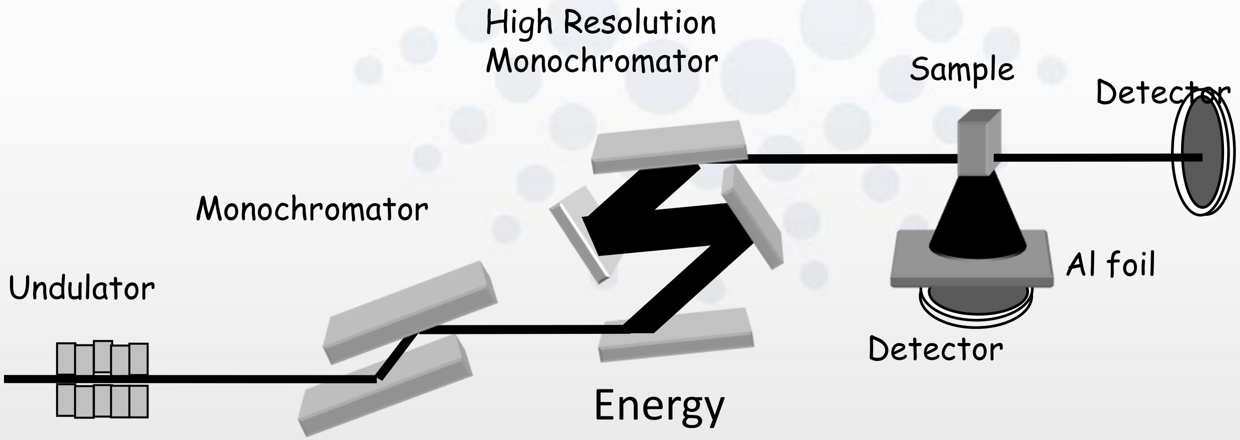
$$I(t) \propto e^{-t/\tau} \cdot \begin{Bmatrix} 1 - 2A_{22} \cdot G_{22}(t) \cdot P_2(\cos\theta) - \\ -\frac{1}{4} A_{44} G_{44}(t) \cdot \left(P_4(\cos\theta) - \frac{1}{24} P_4^{(4)}(\cos\theta)\cos 4\phi\right) \end{Bmatrix}$$

D. H. Ryan and J.M.Cadogan, Hyp. Int. 153 (2004) 43



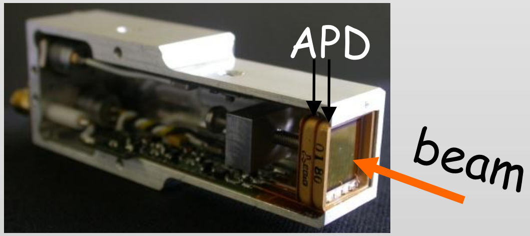


Experimental setup



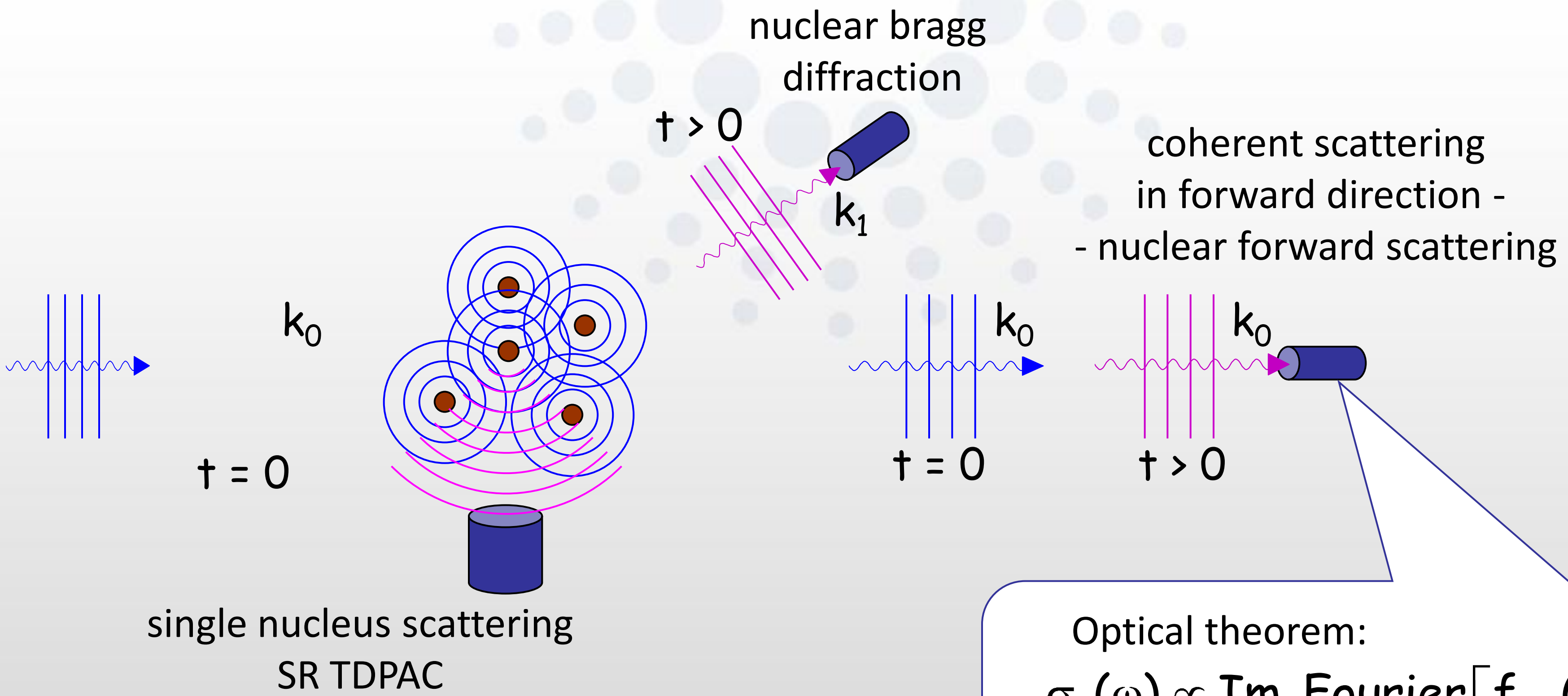
Energy
band width:
1-30 meV

Detector with Si APDs



Dynamical range: ~10^6 ph/s
Time resolution: ~0.1-2 ns

SR TDPAC vs NFS



Optical theorem:

$$\sigma_t(\omega) \propto \mathrm{Im}\ \mathrm{Fourier}\left[f_{fwd}(t)\right]$$

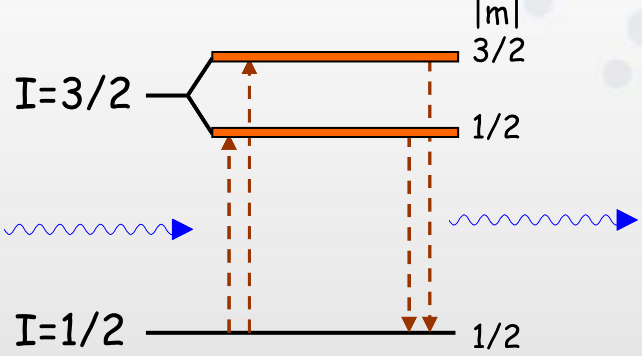
MS total cross-section — NFS amplitude

Differences:

- dependence on the spatial evolution of nuclei
- dependence on the ground nuclear state spin evolution

Hyp. Int.: quadrupole splitting

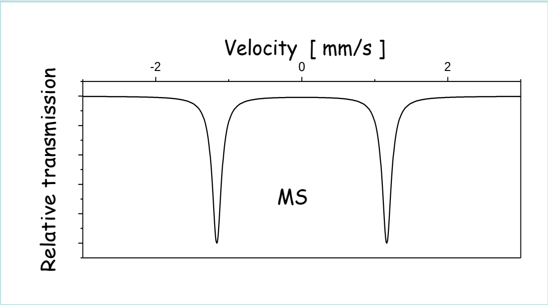
Energy level diagram:
^{57}Fe, quadrupole splitting

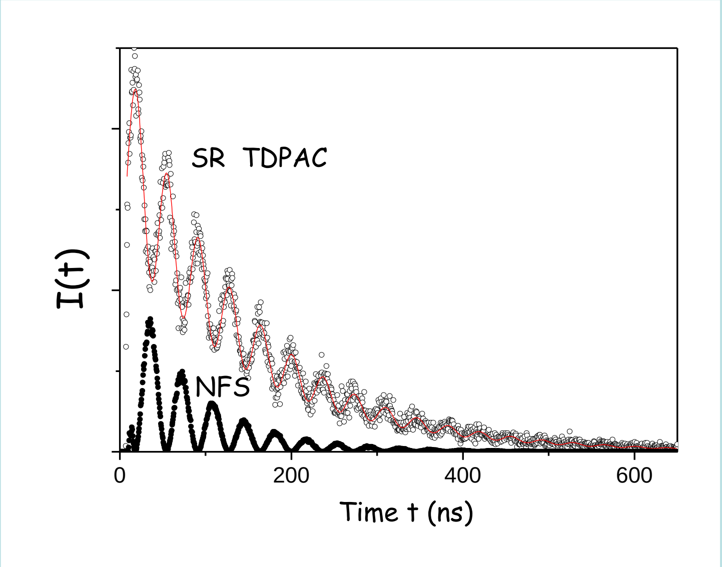


Coherent superposition of the wavelets

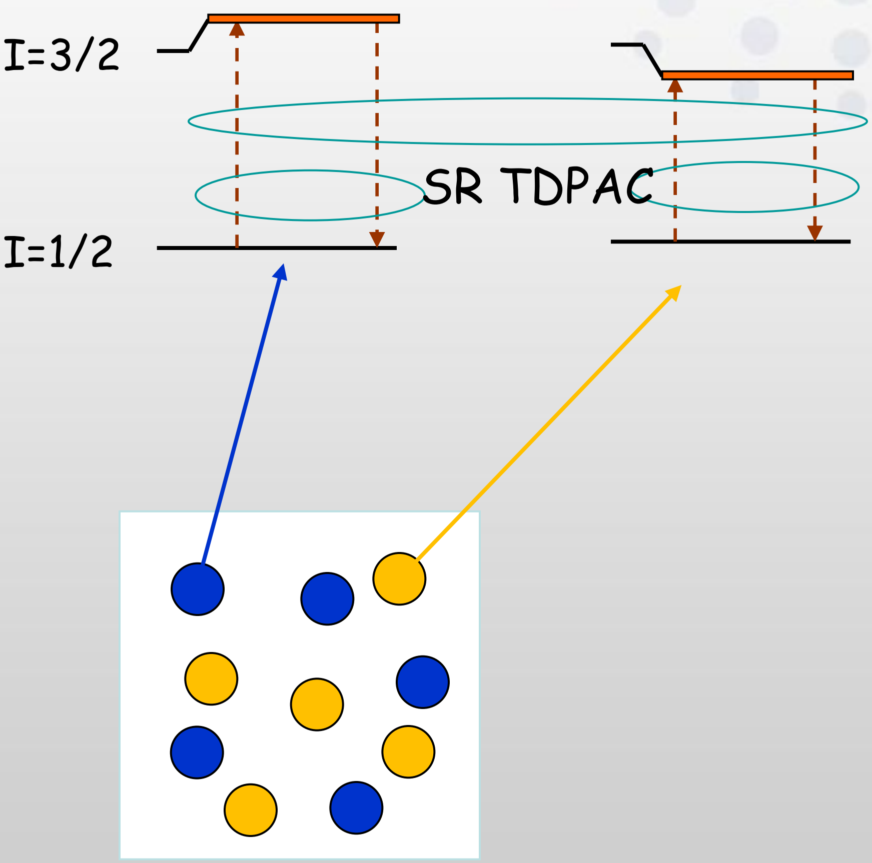
$$E \propto e^{i\Omega t} + e^{-i\Omega t}$$

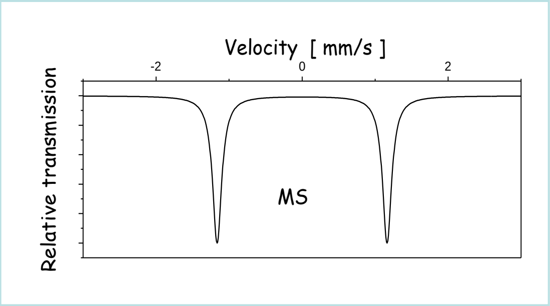
Ferrocene

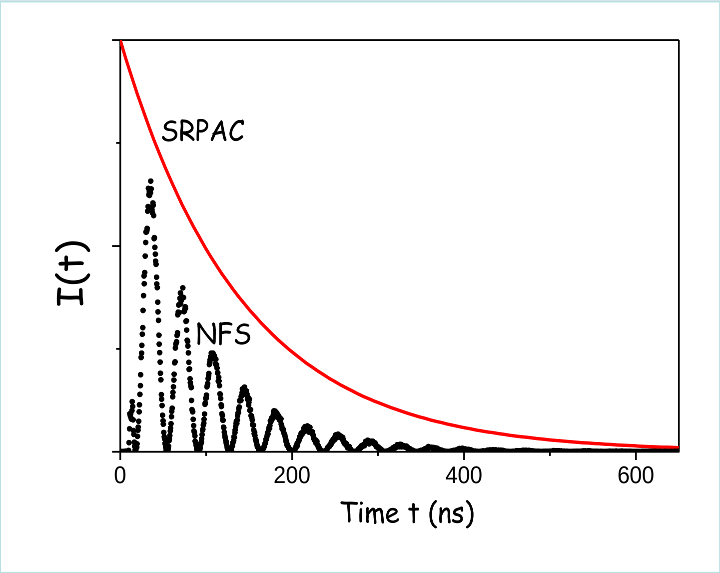




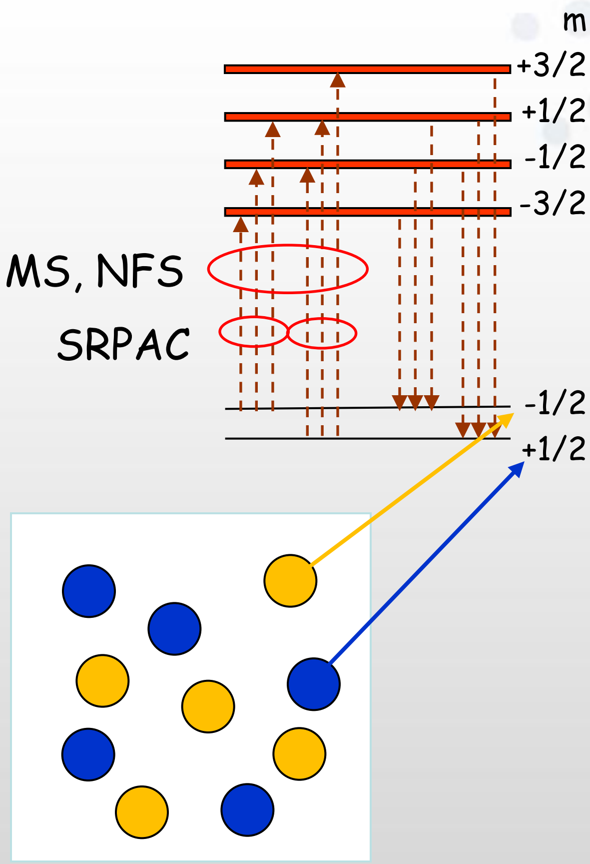
Hyp. Int.: 2 single lines



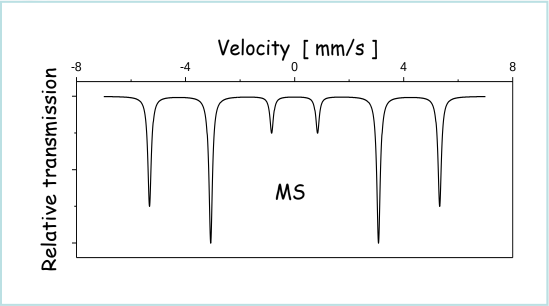


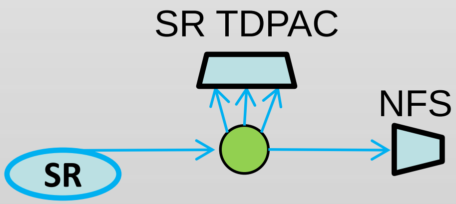


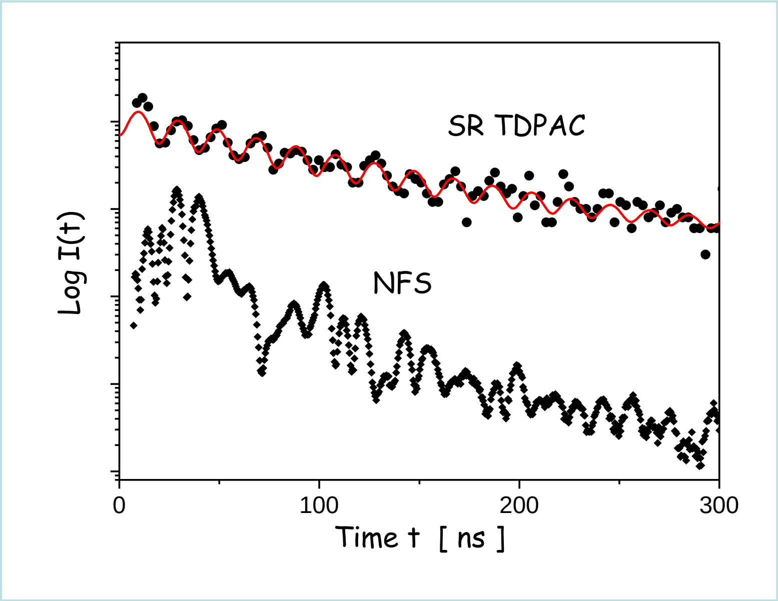
Hyp. Int.: magnetic splitting



α-iron







Applications

- Site-specific phonon dynamics
- Relaxation in glasses
- Study of QI in β-tin

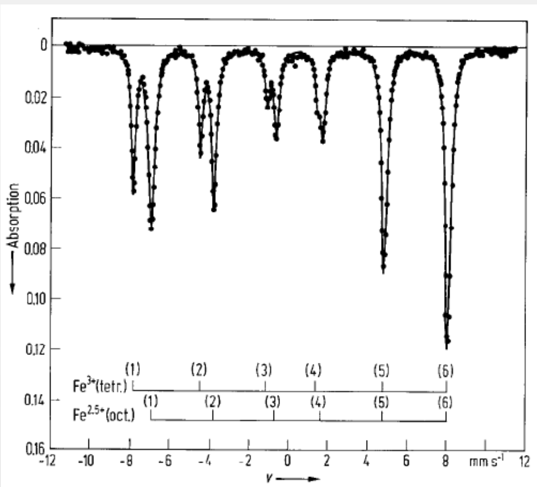
Phonon assisted SR TDPAC

VOLUME 91, NUMBER 18 PHYSICAL REVIEW LETTERS week ending 31 OCTOBER 2003

Site-Specific Phonon Density of States Discerned using Electronic States

Makoto Seto,[1,3] Shinji Kitao,[1] Yasuhiro Kobayashi,[1] Rie Haruki,[1,*] Yoshitaka Yoda,[2]
Takaya Mitsui,[3] and Tatsuo Ishikawa[4]

Weber, H., Hafner, S.S.: Z. Krist. ***133*** *(1971) 331.*

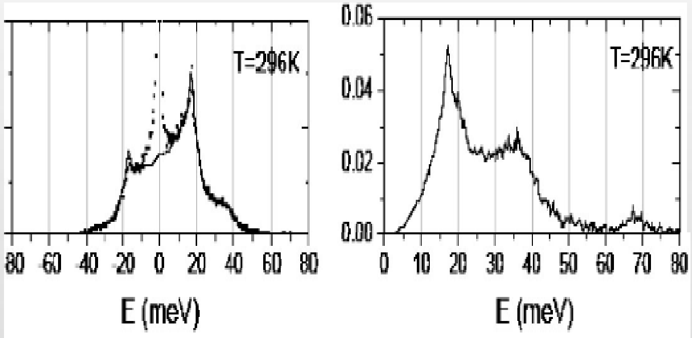


Sample:
Magnetite, Fe_3O_4

A site: tetrahedral, B = 49.0 T

B site: octahedral, B = 46.0 T

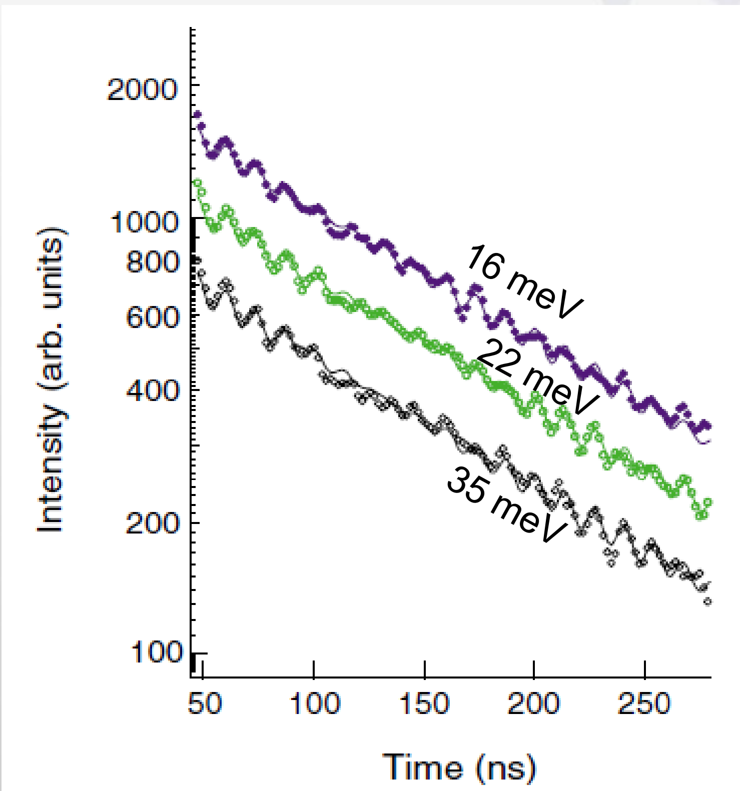
Handke B et al., Phys. Rev. B ***71*** *(2005) 144301*

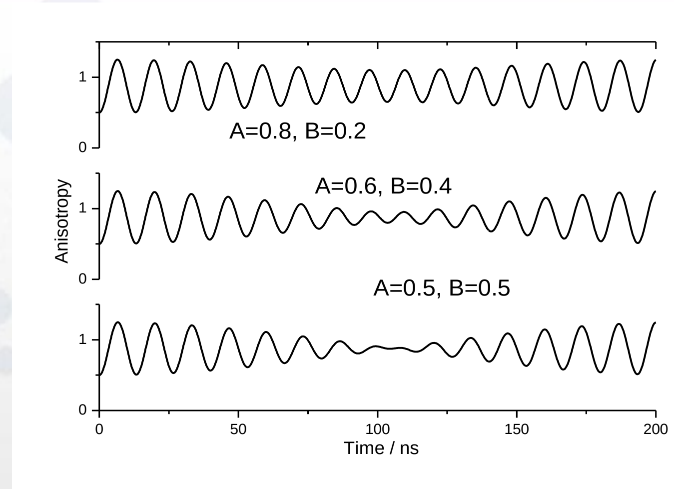


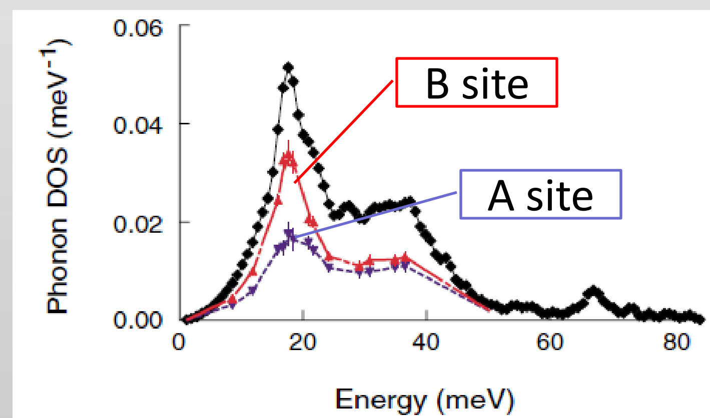
Phonon assisted SR TDPAC

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

$$R(t) = A \cdot (\tfrac{1}{4} + \tfrac{3}{4}\cos 2\omega_A t) + B \cdot (\tfrac{1}{4} + \tfrac{3}{4}\cos 2\omega_B t)$$







Relaxation in glasses

JOURNAL DE PHYSIQUE *Colloque* C6, *supplément au n°* 12, *Tome* 37, *Décembre* 1976, *page* C6-745

MOLECULAR MOTIONS IN A VISCOUS ORGANIC LIQUID : FERROCENE IN COLD BUTYL PHTHALATE (*)

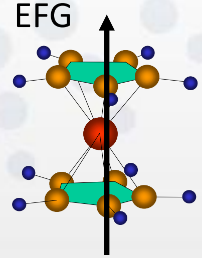
S. L. RUBY and B. J. ZABRANSKY
Argonne National Laboratory, Argonne, Illinois 60439, U. S. A.
and
P. A. FLINN
Carnegie Mellon University, Pittsburgh, Pennsylvania 15213, U. S. A.

Sample:

ferrocene

$Fe(C_5H_5)_2$

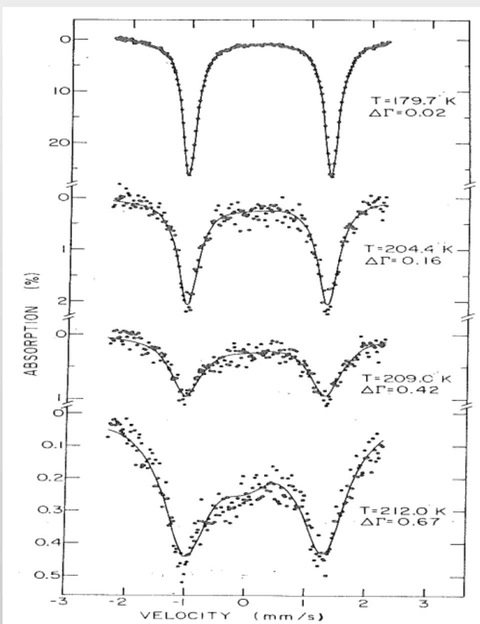
EFG



Glass-former:

Di-butyl phthalate,

T_g = 178 K

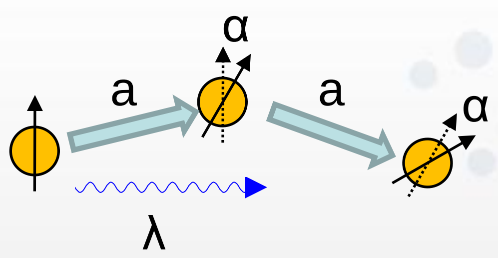


Task:

resolve rotational and translational degrees of motions

Relaxation in glasses

Relaxation process



k – jump rate, sec^{-1}
a – jump distance
α – jump angle
λ – wave length, 0.86 A
R – radius of the molecule, 2 A

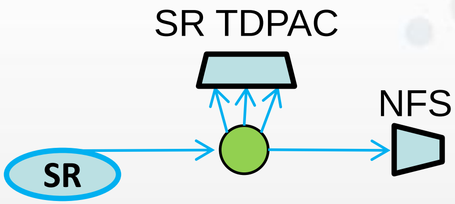
$$\Delta_T = \tfrac{2}{3}\pi^2 \cdot k \cdot \left(\frac{a}{\lambda}\right)^2$$

$$\Delta_R = \tfrac{3}{2} \cdot k \cdot \alpha^2$$

$$\alpha = \sqrt{\tfrac{2}{3}} \cdot \frac{a}{R}$$

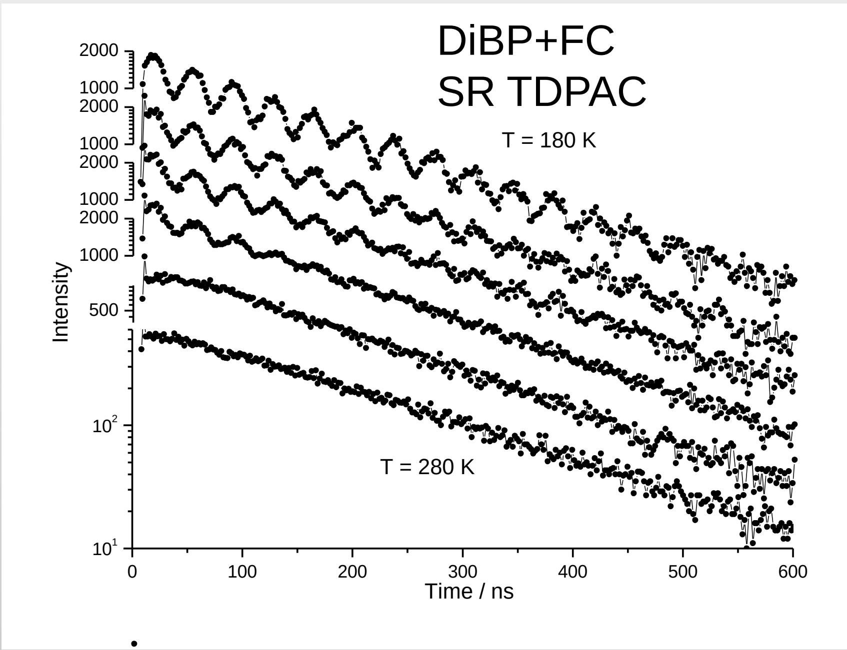
$$\frac{\Delta_R}{\Delta_T} = \frac{3}{2\pi^2} \cdot \left(\frac{\lambda}{R}\right)^2 \rightarrow 0.03$$

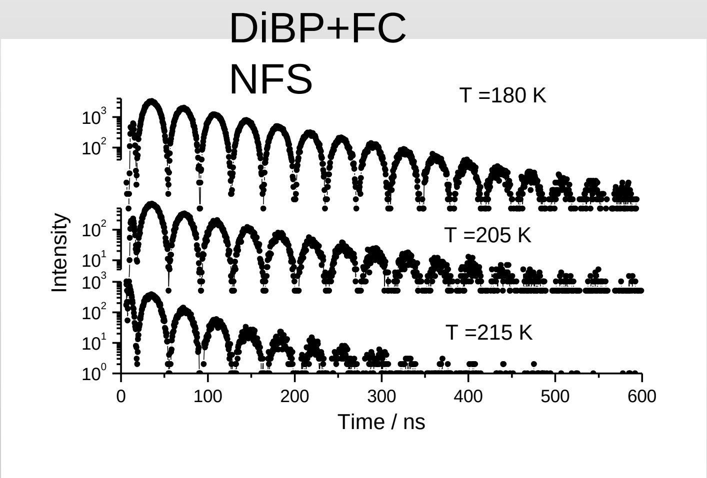
Relaxation in glasses



Sample:

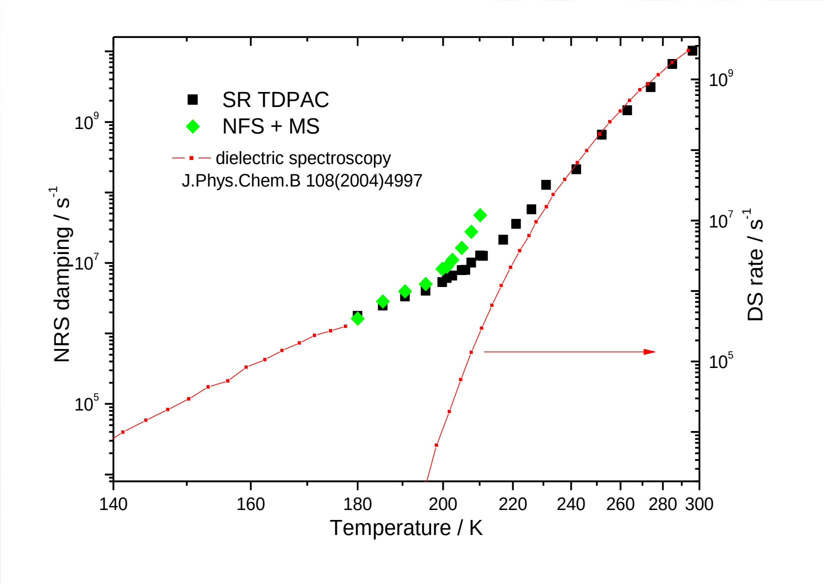
5% ferrocene + Di-butyl phthalate, T_g = 178 K
Di isobytyl phthalate, T_g = 188 K



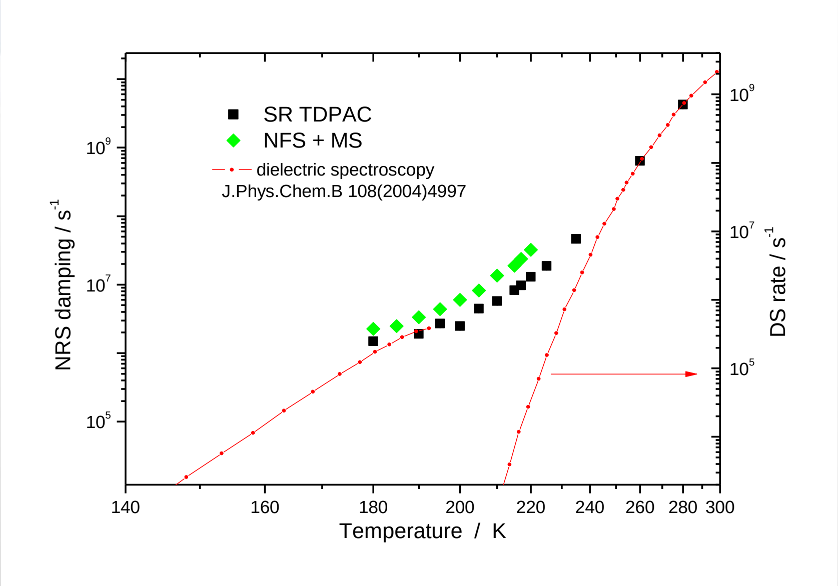


Relaxation in glasses

DBP+FC



DiBP+FC



Conclusions:
The probe reproduces the dynamics of the glass former.
At low T dynamics of the probe follows slow β branch
At low T only molecular rotation is seen in both NFS and SR TDPAC.

Study of β-tin by SR TDPAC

C. Strohm et al., in preparation

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

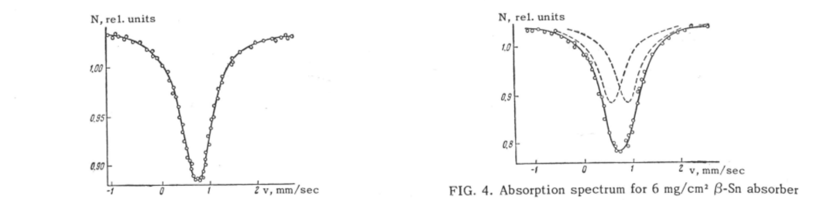
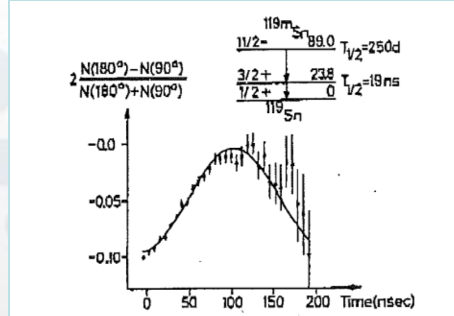


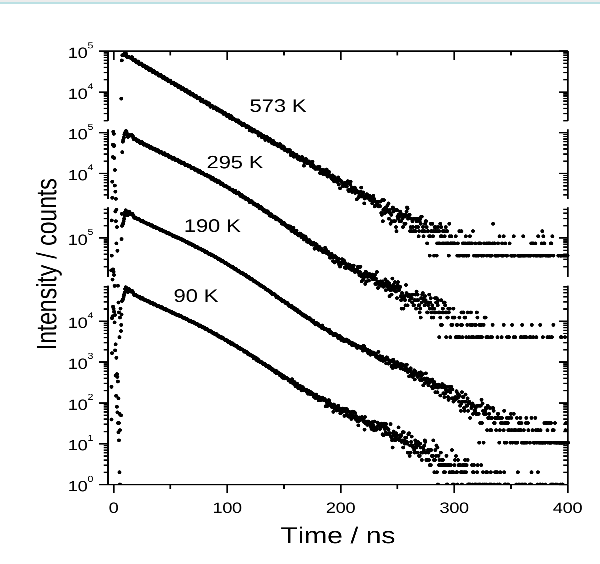
FIG. 3. Absorption spectrum for 22.4 mg/cm² β-Sn absorber at 290°K.

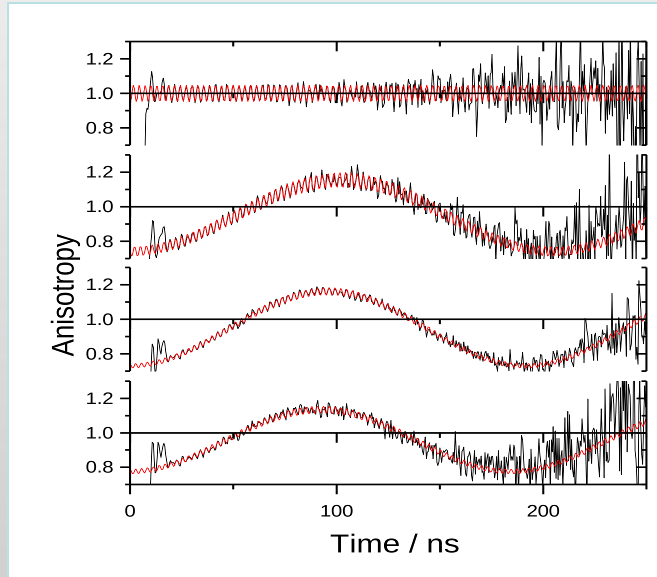
FIG. 4. Absorption spectrum for 6 mg/cm² β-Sn absorber at 77°K.



J.C. Soares et al., Phys.Lett. 45A(1973)465

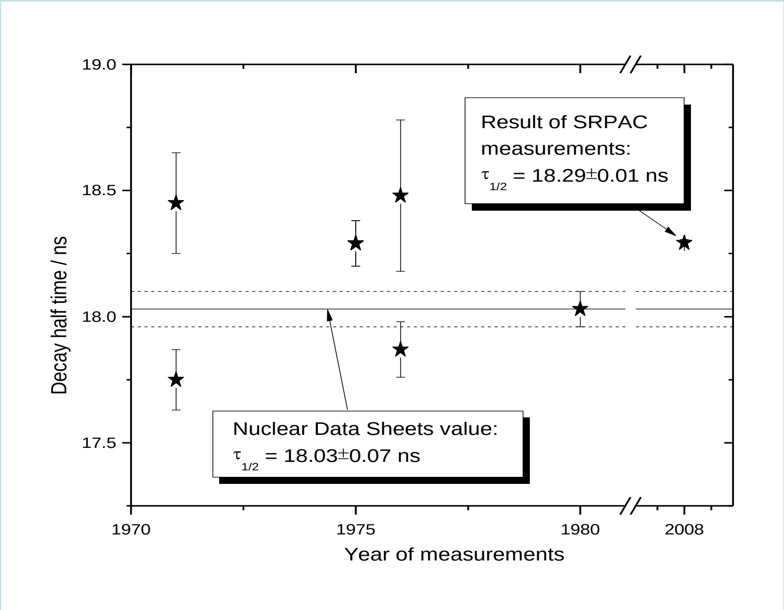
Measurements by SR TDPAC

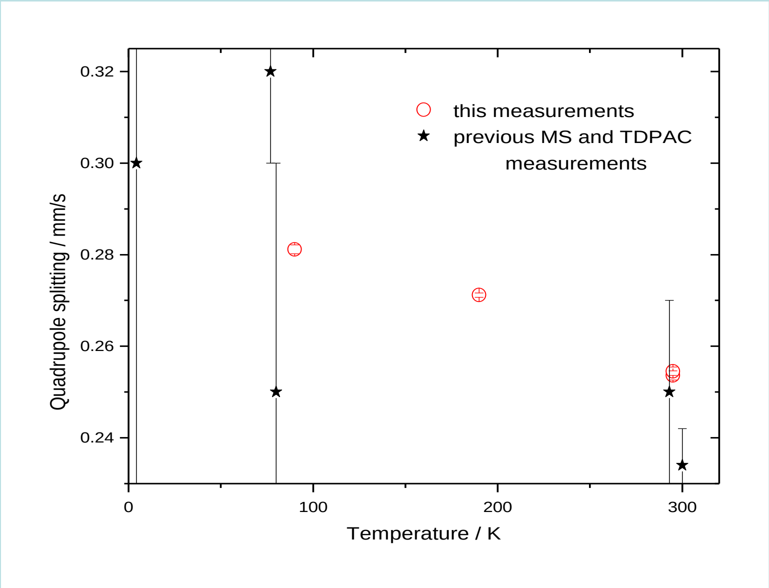




Study of β-tin by SRPAC

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 β-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

Acknowledgment

• U. Van Bürck – TU München, Germany
• G. Smirnov – Kurchatov Institute, Moscow, Russia

Members of ESRF nuclear resonance group:
• R. Rüffer
• A. Chumakov
• T. Asthalter
• C. Strohm
•T. H. Deschaux-Beaume