INTRODUCTION TO MÖSSBAUER SECTION (20 MIN)

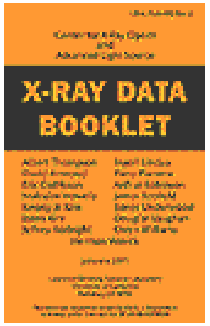
1. Introduction to Mössbauer spectroscopy
   (nuclear resonance scattering):
   - what one can do with scattering of X rays by nuclei
   - how one can do that using SR or XFEL radiation

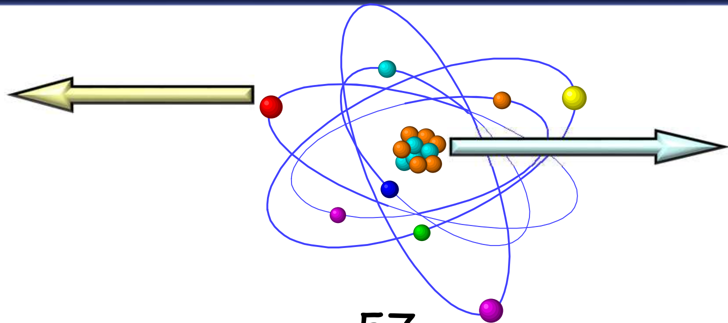
2. New science I: XFELO plus Nuclear Resonance Scattering:
   - *pump-probe experiments with various NRS techniques*
   - enabling new science with $\times 10^3$ higher flux

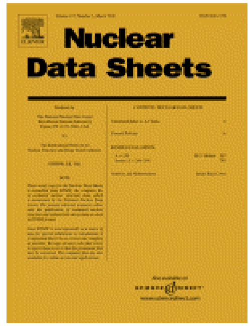
3. New Science II: Multiple mutually coherent nuclear excitation:
   - superradiance: "sorting effect"
   - superradiance: non-linear nuclear optics?

Aleksandr Chumakov, ESRF

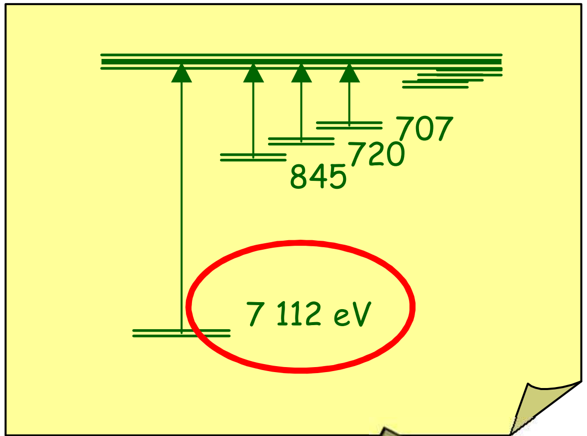
Electronic and nuclear levels

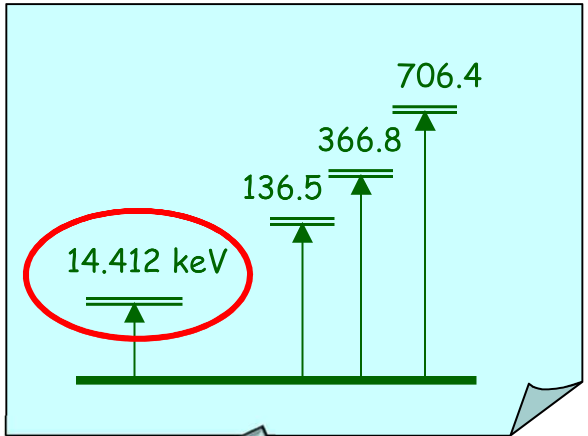






$^{57}_{26}Fe$





$E = 7.112$ keV

$\sigma_e = 26 \times 10^{-20}$ cm^2

$\Gamma \approx 2$ eV

$\tau_0 \approx 0.33$ fs

$$f = f'_e + i f''_e + \frac{g_n}{\Delta E + i\Gamma/2}$$

$E = 14.412$ keV

$\sigma_n = 256 \times 10^{-20}$ cm^2

$\Gamma = 4.7 \times 10^{-9}$ eV

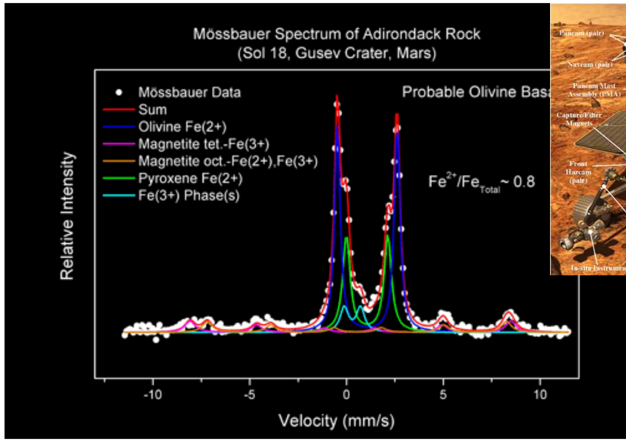
$\tau_0 = 141$ ns

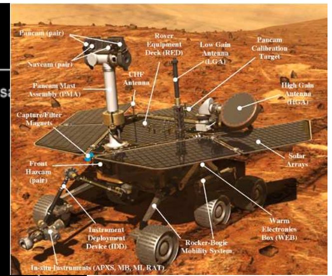
WHAT ONE CAN DO WITH SCATTERING OF X RAYS BY NUCLEI

Narrow resonance: hyperfine spectroscopy

- quadruple splitting:
  - coordination to neighbors
- magnetic splitting:
  - value and direction of magnetic field

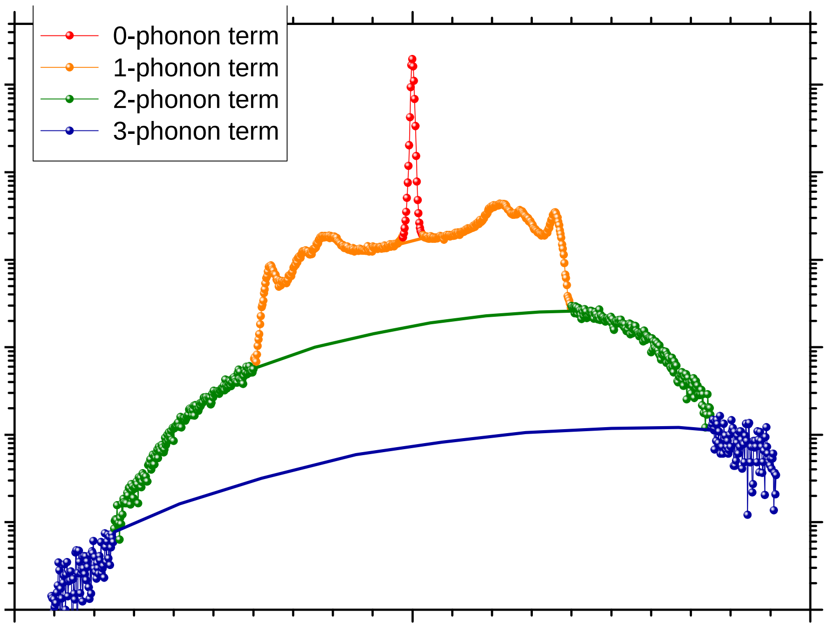
identification of chemical, spin, and magnetic state and corresponding relaxational phenomena





Fixed resonance energy: energy analysis

- nuclear inelastic scattering:
  - partial density of phonon states
- inelastic x-ray scattering:
  - density of phonon states
  - dispersion relations
  - relaxation time

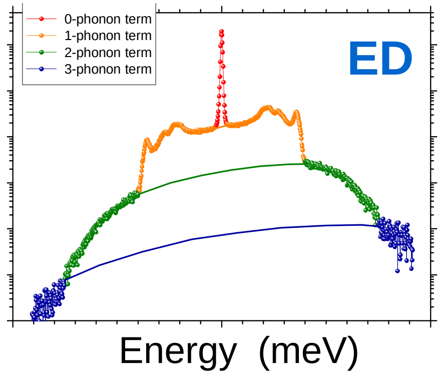


HOW ONE CAN DO THAT USING SR OR XFEL RADIATION

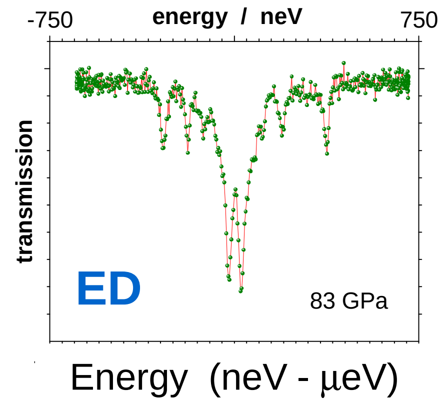
incoherent, for any f_{LM} | coherent, for $f_{LM} > 0$

TD – time differential,
all energies

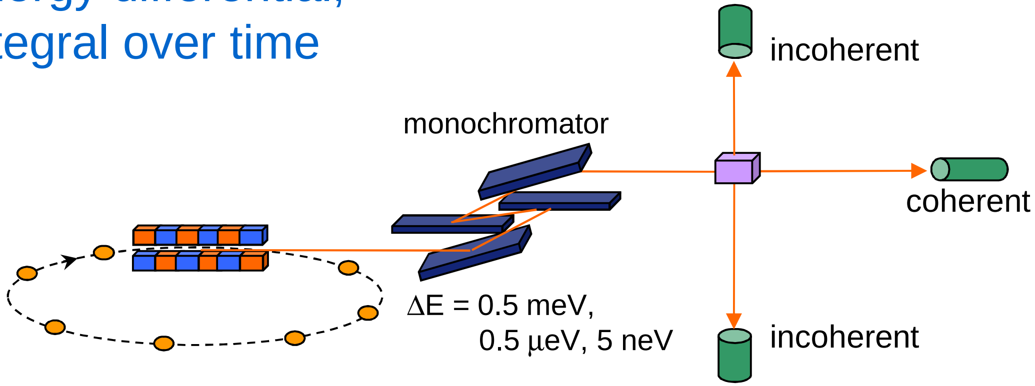
ED – energy differential,
integral over time

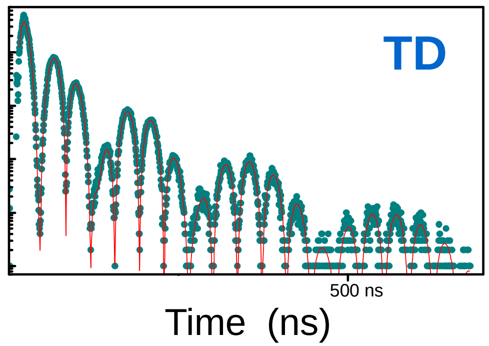


nuclear inelastic scattering

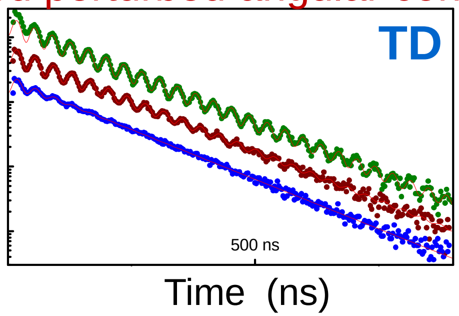


hyperfine spectroscopy
in energy and time domain





hyperfine spectroscopy with
SR-based perturbed angular correlation



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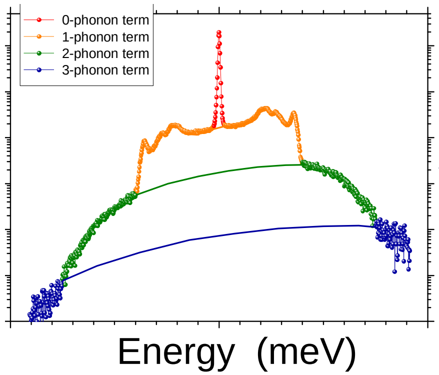
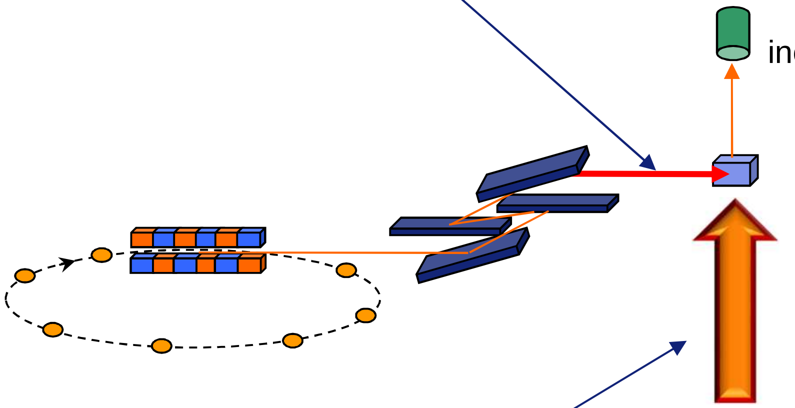
PUMP-PROBE STUDIES: NUCLEAR INELASTIC SCATTERING



Nuclear Inelastic Scattering gives a snapshot of lattice dynamics at the moment of probe pulse and does not depend on evolution of a sample during collecting delayed radiation

Probe: $t = 0$

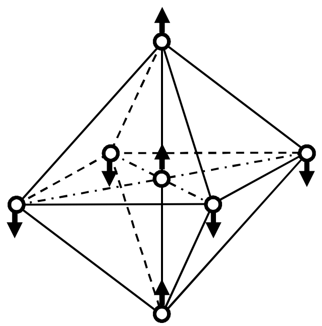


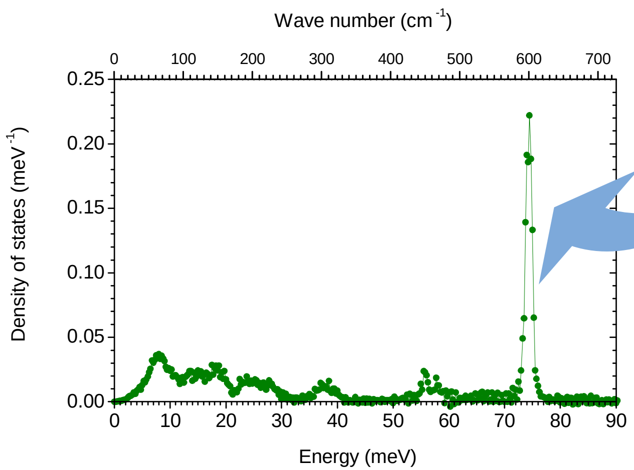
Pump: $t = 0 - \delta$

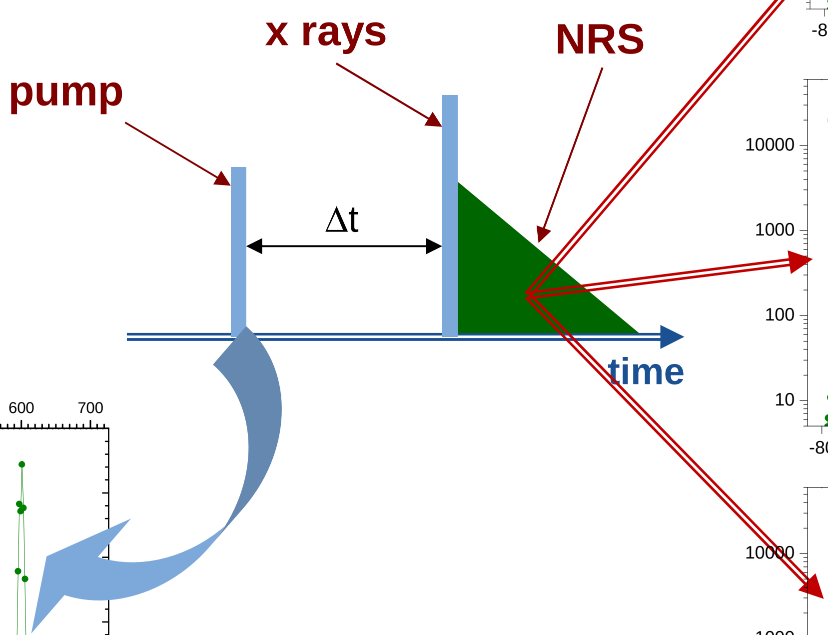
This gives an access to:
heat capacity, heat conductivity
sound velocities
vibrational entropy
identification of melting,
non-equilibrium lattice dynamics
<u>at extreme condition</u>

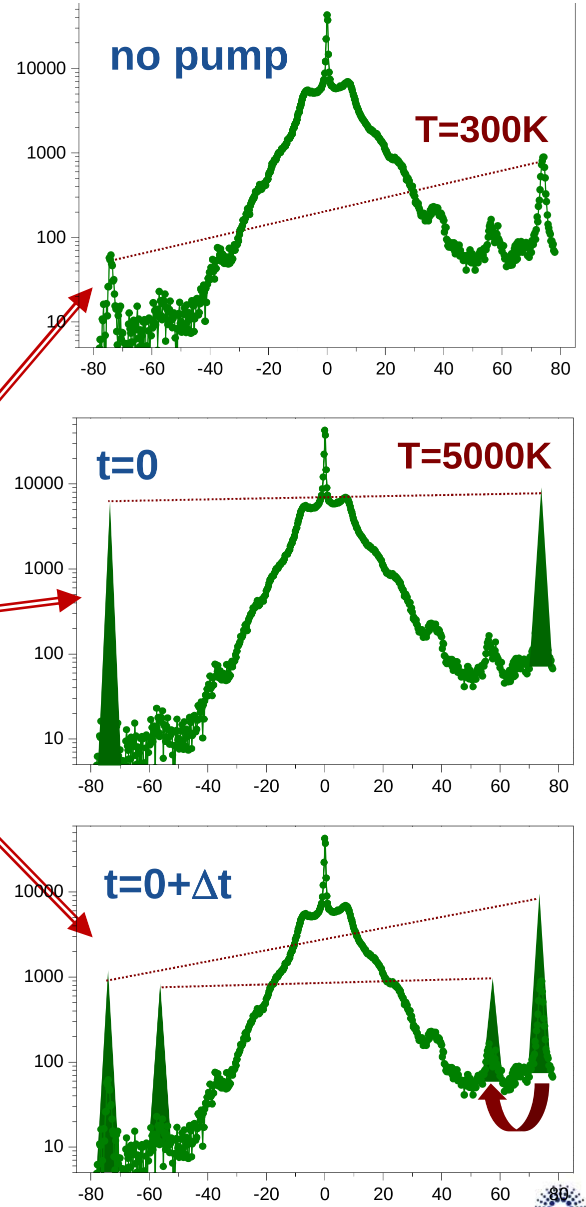
Possible experiment:
non-equilibrium dynamics

$(NH_4)_2MgFe(CN)_6$









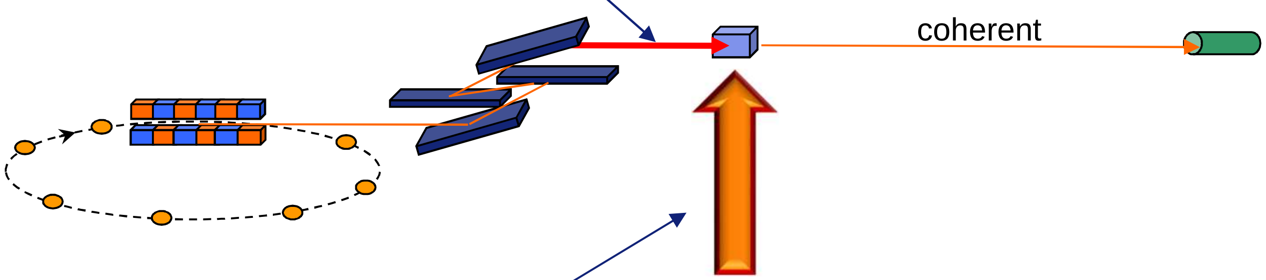
$$\frac{I(+E)}{I(-E)} = \exp\left(\frac{E}{k_B T}\right)$$

PUMP-PROBE STUDIES: COHERENT SCATTERING

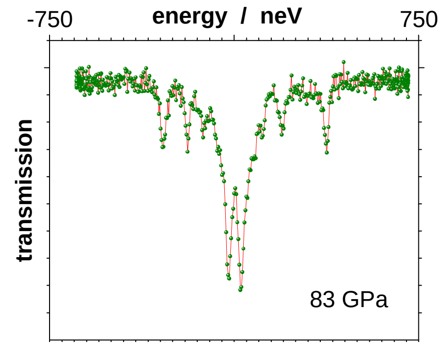
Coherent scattering
(both in time and energy domains)
depends on a sample state at the moment of probe pulse
and on evolution of a sample during collecting radiation

BUT:
if at $t = 0$ $f_{LM}=0$, then no signal
if at $t = 0$ $f_{LM}>0$, then signal exists

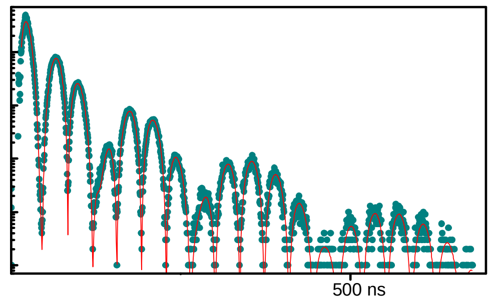
Probe: $t = 0$



Pump: $t = 0 - \delta$



Energy (neV - μeV)



Time (ns)

This may give an access to:
identification of melting,
time-resolved hyperfine spectroscopy
e.g., *non-equilibrium spin states*

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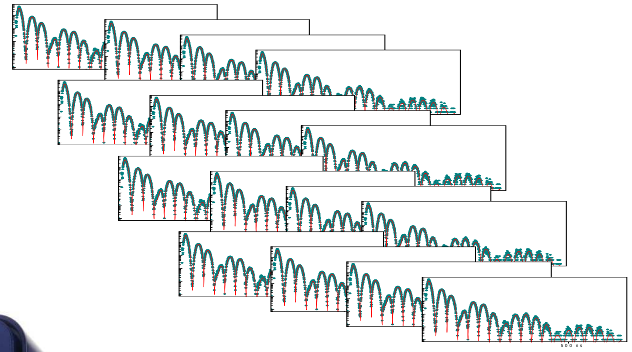
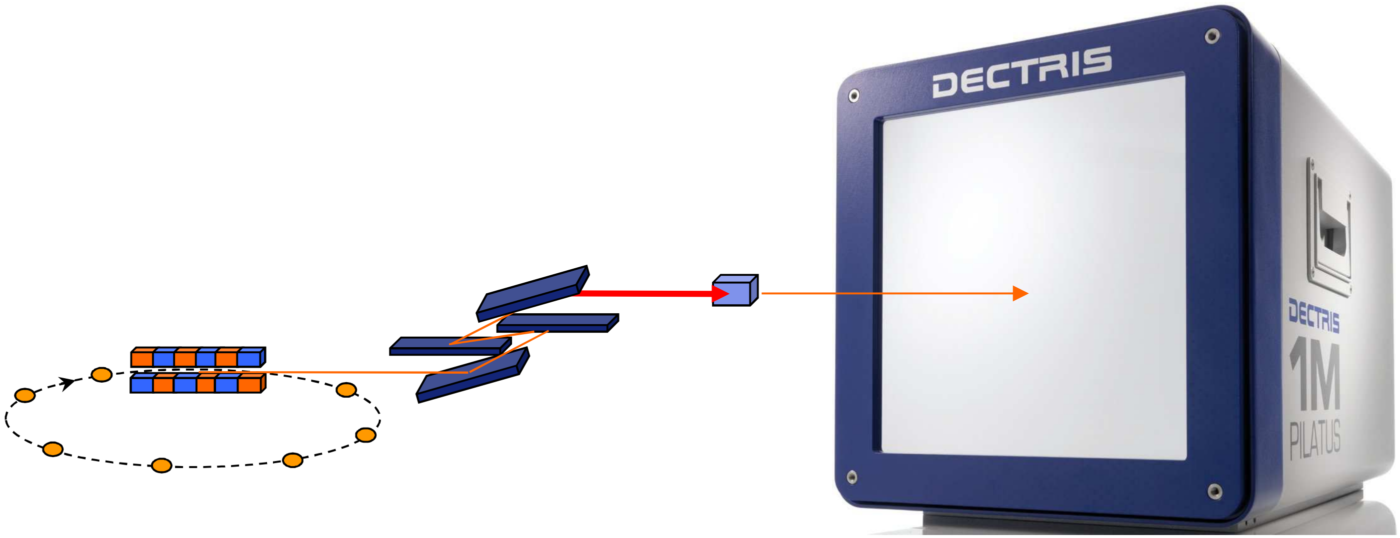
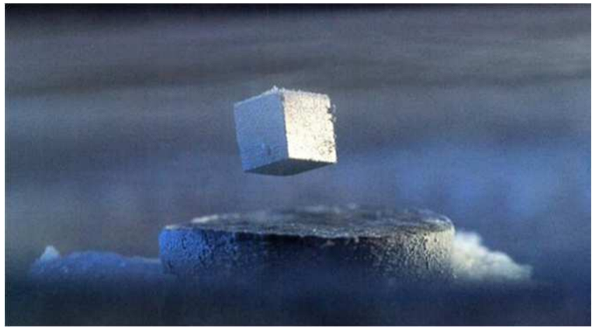
2D – SPECTROSCOPY:

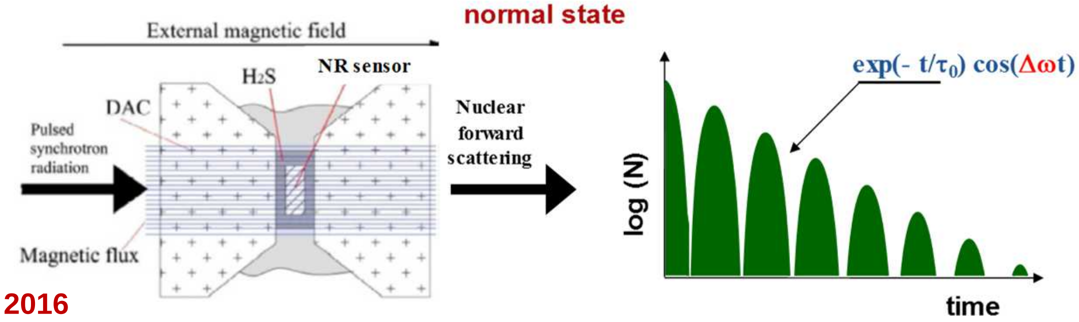




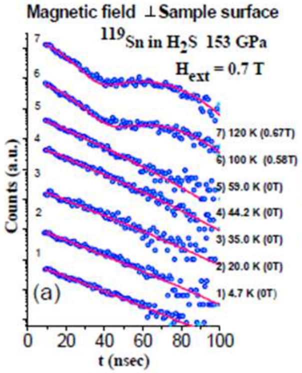
- time-resolved 2D detector
- 100:1 magnification of sample image

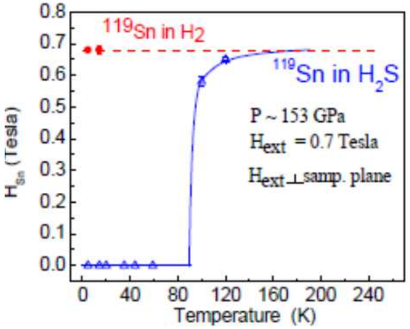


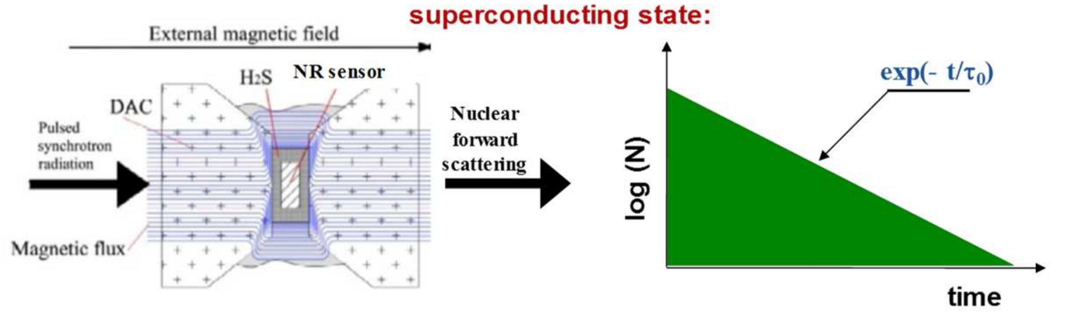
Superconductivity: Meissner effect



Troyan *et al.*, Science, 18 March 2016

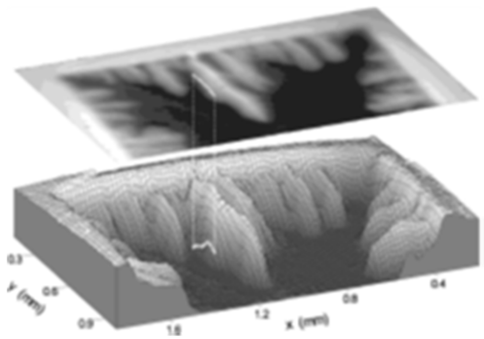


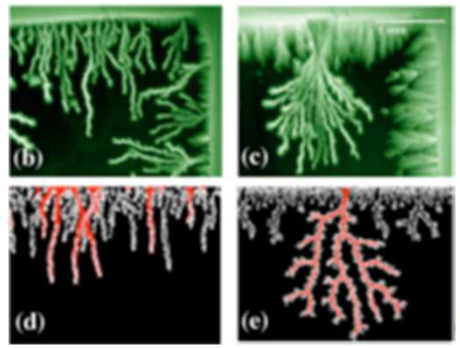




spatial distribution of superconducting phases: disentangling bulk and "lamella" superconductivity,

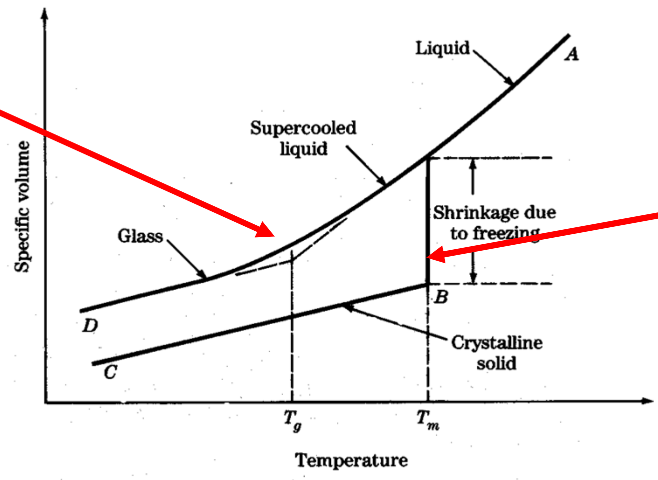
visualization of the vortex structure.





second order transition?

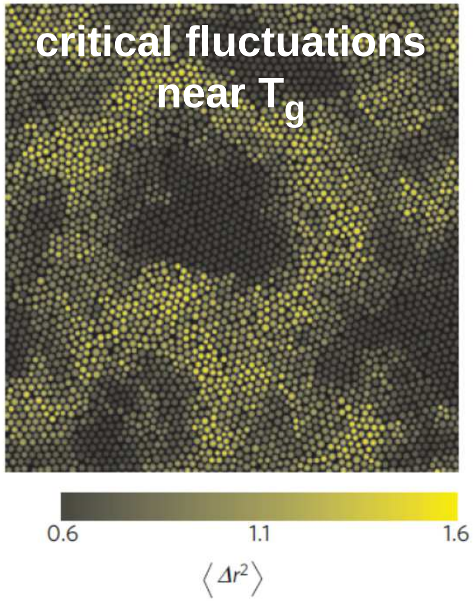
first order transition in *cooperative dynamics* ?!



first order phase transition

Tanaka *et al.*, Nature Materials 2010

fluctuations in *m.s.d.*



Glass transition: dynamical heterogeneities

characterized by four-point correlation G_4(r,t) or susceptibility χ_4(t) functions

no experimental methods exist for real systems

totally new experiment:
quasi-elastic small-angle nuclear resonance scattering

effective thickness: $T = \sigma_0 n d f_{el}$

***fast* scattering:**
Debye-Waller factor

$$f_{DW} = exp(-\Delta k^2 \Delta x^2)$$

not sensitive to m.s.d.
for small angles

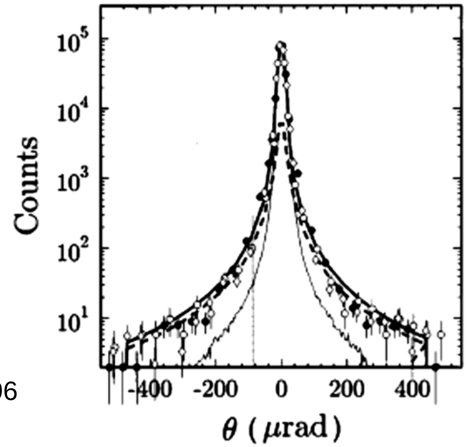
***slow* scattering:**
Lamb-Mössbauer factor

$$f_{LM} = exp(-k_0^2 \Delta x^2)$$

small-angle scattering
from inhomogeneous m.s.d.

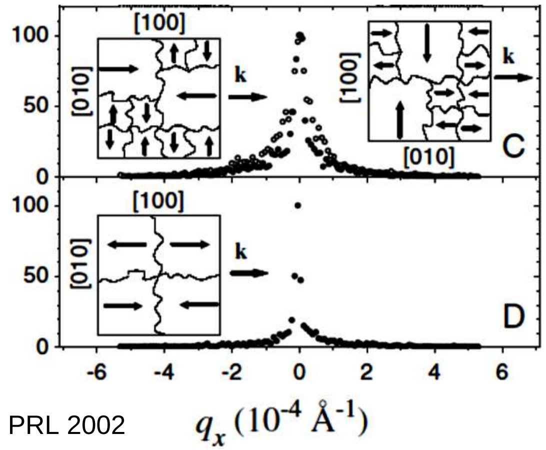
elastic small-angle nuclear resonance scattering:

magnetic domains
in α-iron



Shvyd'ko *et al.*, PRL 1996

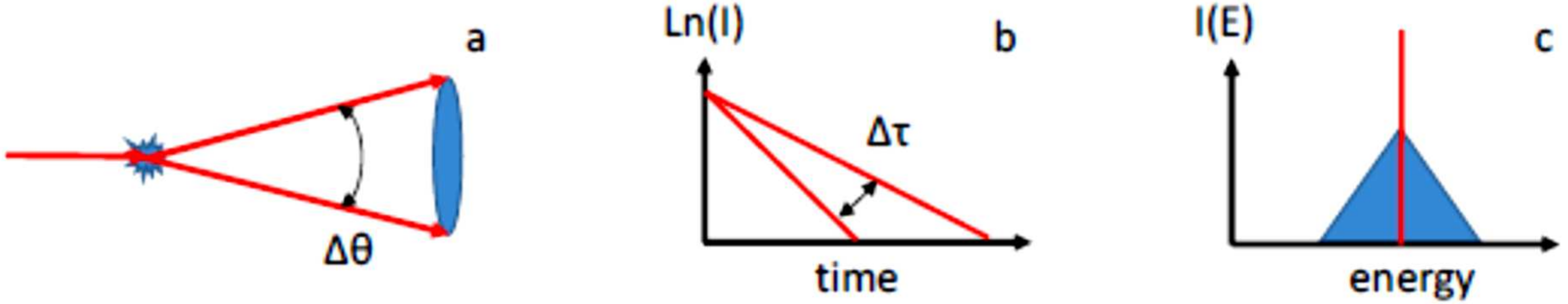
magnetic domains
in a Fe/Cr
multilayer



Nagy *et al.*, PRL 2002

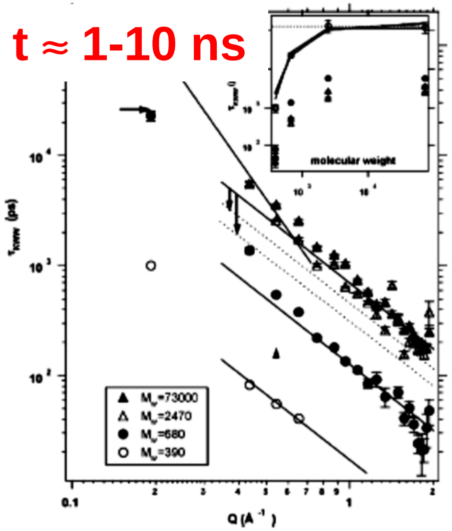
dynamical heterogeneities

complete characterization of dynamic heterogeneities
accessing the correlation length and the correlation time of atomic mobility

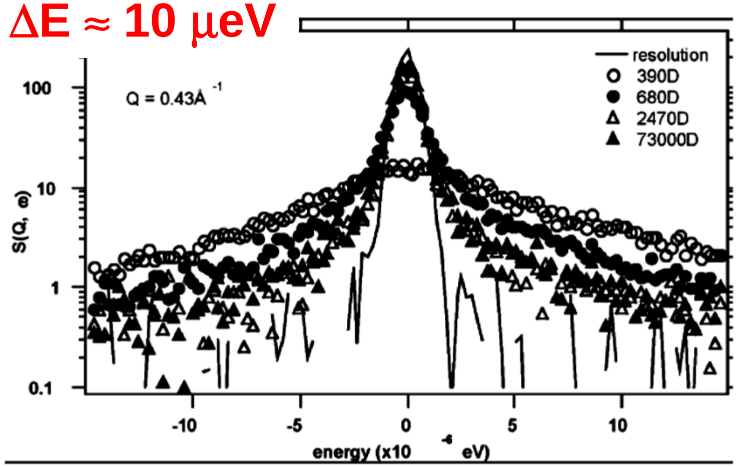


$$\Delta\theta = \frac{\lambda}{10\,nm} \approx mrad$$

t ≈ 1-10 ns



ΔE ≈ 10 μeV



QENS in PB
for various MW

Frick *et al.*, Chem.Phys. 2003

To access the relaxation energy range: ~<u>10 μeV resolution</u>

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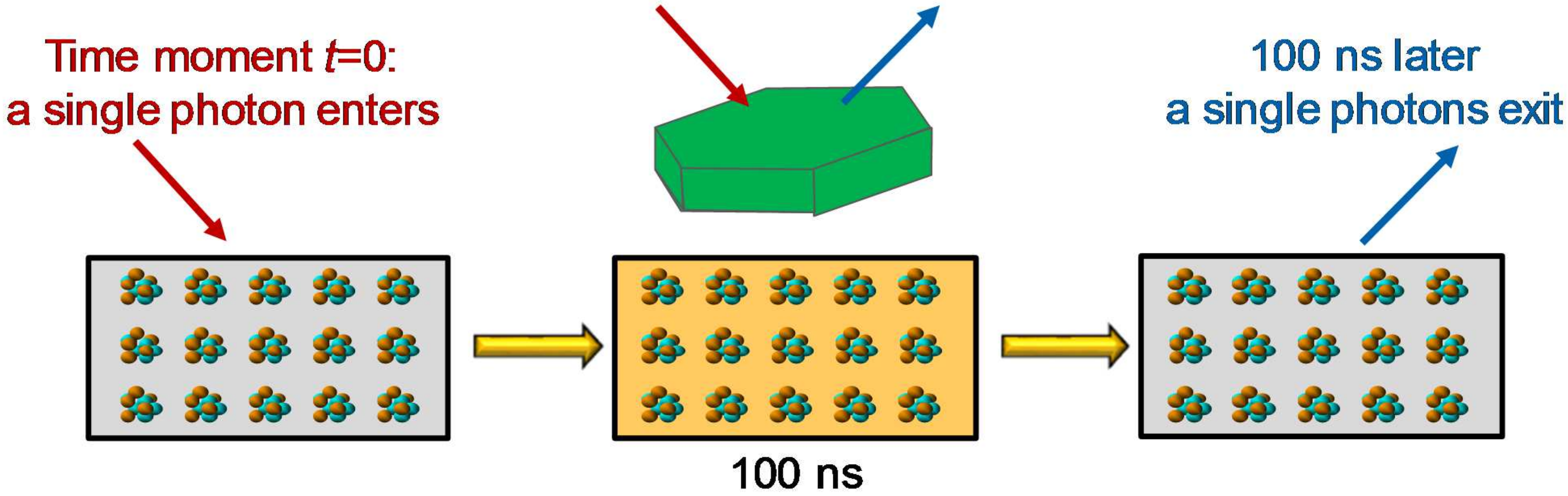
MULTIPLE MUTUALLY COHERENT NUCLEAR EXCITATION

Question:
where was the photon during 100 ns?
(how many nuclei were excited?)

Answer:
it created compound excited state,
all nuclei (within transverse coherence region) were excited

Retarded nuclear diffraction:



Question:

What will happen if we excite the same compound excited state (i.e., the same nuclei) several times by mutually coherent excitations?

SUPERRADIANCE

Phys. Rev. **93**, 99 (1954).

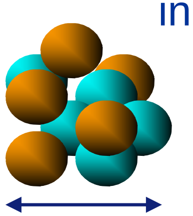
Coherence in Spontaneous Radiation Processes

R. H. Dicke
Palmer Physical Laboratory, Princeton University, Princeton, New Jersey
(Received August 25, 1953)

3760 citations

By considering a radiating gas as a single quantum-mechanical system, energy levels corresponding to certain correlations between individual molecules are described. Spontaneous emission of radiation in a transition between two such levels leads to the emission of coherent radiation. The discussion is limited first to a gas of dimension small compared with a wavelength. Spontaneous radiation rates and natural line breadths are calculated. For a gas of large extent the effect of photon recoil momentum on coherence is calculated. The effect of a radiation pulse in exciting "super-radiant" states is discussed. The angular correlation between successive photons spontaneously emitted by a gas initially in thermal equilibrium is calculated.

simplest case:
many excited atoms
in a single point



d << λ

Superradiance:
N-times acceleration
of the initial decay rate
for *N*-photon excitations

Phys. Rev. A **3**, 1735 (1971).

Superradiance*

Nicholas E. Rehler† and Joseph H. Eberly
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627
(Received 2 October 1970)

We introduce a straightforward quantum-electrodynamic approach to the problem of superradiant spontaneous emission from a system of two-level atoms, and also discuss two classical superradiant systems. The same physical concepts underlie our treatment of both the quantum and classical cases. Explicit expressions are found which describe the time evolution and directional character of the radiated power. Our approach applies to an arbitrary number of atoms which are confined to a region with linear dimensions which may be large compared with the mean wavelength of the emitted radiation. The radiative decay half-life of a large many-atom system is found to be much shorter than the natural lifetime of a single atom, but not as short as the half-life of a small atomic system with the same number of atoms. The far-field radiation pattern appropriate to the case of a circular cylinder of emitters which have been excited by a plane-wave pulse is considered in detail. Polar plots are presented of the radiation patterns produced when the atoms are excited by a plane-wave pulse. Exciting pulses traveling in directions both parallel and at an angle of 10° to the axis of the cylinder are considered. A discussion and generalization of Dicke's "coherence-brightening" criterion is given.

SUMMARY FOR MULTIPLE NUCLEAR EXCITATIONS:

Nuclear resonance scattering of XFEL radiation is dominated by multiple nuclear excitations.

N-times acceleration of the initial decay for multiple N-photon excitations exists.

At least for small N, it can be described as a sequence of photon sorting (1st photon by definition must come faster than in average).

Possibly, there is an additional acceleration of the initial decay beyond the "sorting" effect. It starts to be visible for N>10, and it is better seen for N>50.

If so, then this seems to be the manifestation of non-linear nuclear optics (dependence of nuclear refraction index on intensity).

SUMMARY FOR INTRODUCTION TO MÖSSBAUER SECTION:

Nuclear resonance scattering at XFELO source promises to enable new science by:

1. **Time-resolved (pump-probe) studies**
   (for inelastic scattering, the applications are straightforward,
   for coherent channels –have to be considered case-by-case)

2. **By the three orders of magnitude higher spectral density.**
   Main promises are in spatial resolution and energy resolution

A particularly interesting application is a non-linear nuclear optics